University of Nottingham
UK | CHINA | MALAYSIA

Hydrothermal carbonisation of digestate: An investigation into the technical design, economic feasibility and future potential of the process.

Sarah Farthing

A thesis submitted to the University of Nottingham for the degree of Doctor of Philosophy

December 2020

Abstract

Hydrothermal carbonisation (HTC) is an emerging technology used for the treatment of wet biomass. The process is carried out in an aqueous environment and so there are large thermal energy savings available for wet wastes that would ordinarily require drying prior to conventional thermochemical processes. HTC converts wet biomass into a solid product known as hydrochar, a process liquid and a small amount of CO_2-rich process gas. The relative yields of the hydrochar and process liquid are influenced by process conditions. The products have potential markets as a fuel, fertiliser and/or for carbon sequestration. Currently, there are a number of industrial-scale HTC plants, but these typically operate under specific contractual agreements with local authorities or generators of problematic waste streams. In order to understand the potential of HTC in the wider market, there is the need for a robust and comprehensive analysis of the technical and economic performance of HTC. This will identify the conditions at which HTC is most feasible and allow for targeted support, development and deployment of the technology.

This work comprised of an experimental analysis of the influence of HTC process conditions using various feedstocks, the development of a fully flexible process model validated by industrial data, and a detailed economic assessment and case study based in the UK. Digestate from the anaerobic digestion (AD) process was selected as the feedstock of focus for this work. In 2019 over 12.5 million tonnes of feedstock were processed with AD in the UK, generating 10.9 million tonnes of digestate by-product, equivalent to 500,000 tonnes on a dry basis. AD capacity and therefore digestate generation is increasing year on year, but issues remain for its use or disposal of digestate. The high moisture content, variable composition and restrictions on nutrient levels complicates and limits its spreading to agricultural land. There is a need for energy efficient, environmentally sound and economically viable treatment process. Current digestate management costs in the region of £16/tonne of digestate fibre, with a solids content of 25 wt.%, and it has been suggested that HTC could offer an overall lower-cost solution.

This study is the first economic model of HTC using digestate as a feedstock. Three different digestate feedstocks were used experimentally: mixed food waste (waste-based), livestock slurry and silage (farm-based) and maize (crop-based). The hydrochar compositions reflected the feedstock used. The waste-based and farm-based hydrochars contained high levels of ash, often in the region of 30–50 wt.%, meaning they were deemed as unsuitable as solid fuel products. However, the hydrochar from the crop-based digestate had a carbon content of over 50 wt.% and ash content of below 10 wt.%, similar to bituminous coal. The waste-based and farm-based digestates and their resulting hydrochars had higher nutrient contents and so their

use as a fertiliser or growing media was investigated. The results showed that they had promise in these areas, but more conclusive studies are required to understand the business case, especially concerning the presence of phytotoxic compounds and the long-term stability of the carbon. HTC experiments were carried out at a range of process conditions and it was found that temperature had the largest effect on the products, agreeing with existing literature results. A higher HTC temperature (of up to 250 °C) increased the degree of carbonisation, that is, an increased carbon content of the hydrochar but lower mass yield. The mass yield of hydrochar was typically measured as between 45 and 65 wt.% across all conditions. An increase in the initial feedstock moisture content was also found to increase the degree of carbonisation, although to a lesser degree than temperature. An increase in residence time was found to be statistically insignificant in nearly all cases.

A process model was developed based on the experimental results and industrial HTC plant designs. This process model is the most detailed and accurate publicly available HTC model to date; it was designed with flexibility in mind along with industrial insights. A 2-reactor HTC plant treating 41,600 tonnes of digestate fibre with a solids content of 25 wt.% was found to require up to 1,000 MWh/year of electrical energy and 10,000 MWh/year of thermal energy. The capital expenditure (CAPEX) of a plant of this size came to below £5.8 million and the operating expenditure (OPEX) was in the region of £1.2 million. A 2-rector plant was found to be the best in terms of the trade-off between economies of scale and costs of transporting feedstock to site. The model results were validated with industrial data and were found to be in good agreement, offering a large improvement in data quality over what is currently publicly available in literature. There are two main revenue streams for a HTC plant treating digestate: the sale of hydrochar and the avoidance of digestate management costs. The sale price depended on the hydrochar properties. Crop-based hydrochar could expect to sell for up to £181/tonne as a solid fuel, whereas the hydrochar from waste-based and farm-based digestates could expect to be sold at £64–79/tonne as a fertiliser or £89–134/tonne in carbon sequestration. While there are a number of variables, in general, the farm-based and crop-based digestate showed better performance, with a payback period for the HTC plant of roughly 14 years or fewer. However, the HTC of waste-based digestate resulted in a payback period in excess of 20 years. A few lignocellulosic feedstocks were fed into the process and economic model for comparison. While they produced hydrochar of higher quality and yield, the high feedstock cost resulted in an economically unfeasible HTC model. With a fixed cost saving of avoiding current digestate fibre management practices of £16/tonne, the hydrochar sale price would need to be £255/tonne, £200/tonne or £190/tonne for the waste-based, farm-based and crop-based digestates, respectively, to achieve a pay-back period of 10 years. Alternatively, by fixing the hydrochar sale price at £150/tonne, the digestate savings would

need to be £30/tonne, £26/tonne or £25/tonne of digestate fibre for a payback period of 10 years for the waste-based, farm-based and crop-based feedstocks, respectively. Ultimately, the HTC of farm-based and crop-based digestate showed promise both technically and economically, and so it is suggested as a possible treatment route in the UK. The waste-based digestate results were not as favourable. However, looking to the future, waste-based digestate has inherent problems with low quality and plastic contaminant content, both of which HTC can handle. Therefore, of the types of digestate considered here, its management costs are predicted to be the largest and most likely to increase. It is expected that under certain conditions and policy changes, it could become economically feasible in time. This study is the first to evaluate the techno-economics of the HTC of various digestate streams in the UK and set out the limits that must be met for it to be suggested as a treatment process.

Acknowledgements

Firstly, I would like to thank my supervisors, Dr Jon McKechnie and Professor Colin Snape for their support and academic guidance throughout my PhD. I would also like to thank the Faculty of Engineering at the University of Nottingham for all the resources, training and provisions. A particular mention goes to Clement Uguna and Robert Stirling for training and assisting me with the laboratory work in this project. Thank you to the various famers and other students for supplying the experimental feedstocks I studied.

I would like to acknowledge the Engineering and Physical Sciences Research Council (EPSRC) for their funding of this project through the Centre for Doctoral Training (CDT) in Sustainable Chemistry. The CDT management team provided support, encouragement and so many brilliant opportunities during my time as a PhD student. Without them, I would not have been able to present at or engage in many conferences, courses and other activities. It was through the CDT that I was able to spend two months with Ingelia in Valencia, who I would like to extend my thanks to. During my time there, I gained a better understanding of the development of industrial hydrothermal carbonisation and was able to visit their plant myself.

My time doing this PhD would not have been the same without the support and friendship from the other CDT students, especially Cohort 3. You have been an excellent team to be part of since day one and I have loved spending this time with you all. The LCA research group have also been brilliant to work alongside. Thank you for the for the critical reviews of my work, laughter and caring for my office plants when I was away. Ultimately, I would like to thank all those I'm closest to, those who have encouraged me and shown me such love during all the highs and lows. Thank you to my parents, Alice, Bruno, Tom, Saskia, Neelam, Jenny, Lenny, Maisie, Woolly, Schnecke, my wonderful Church family and everybody else. And finally, I thank God, for His unfailing love and for being my Rock throughout it all.

Contents

List of Tables

List of Figures

1 Project Introduction

1.1 Overview

Hydrothermal carbonisation is a promising new technology for the treatment of wet, waste biomass and has gained increased interest over the recent years. Indeed, several industrial-scale HTC plants now exist in Europe and Asia. However, these plants are generally operating based on a specific, contractable agreement with a certain waste generator or local authority. There remains a lack of understanding of the feasibility of HTC as a general waste treatment. As interest in waste treatment technologies increases, there are calls for more informed decision making, particularly in policy and implementation strategies [1]. Examples such as the 'cash for ash' scandal in Northern Ireland, resulting from mismanagement of the Renewable Heat Incentive, have been highlighted in order to avoid similar failures in policy making in the future [2]. HTC is capable of processing a wide variety of feedstocks at a range of conditions which ultimately determines the product yields and characteristics. It is a technology with many options and opportunities, although there are significant trade-offs between performance and cost. Due to these multiple factors, assessing the viability and optimisation of HTC on an industrial scale is complex. The purpose of this work is to develop a generic methodology for assessing the technical and economic feasibility of HTC at industrial scale, in order to make more informed decisions about its future deployment.

A range of experiments at different process conditions were conducted and the yields and characteristics of the products were measured. From these results, a process model was created, predicting the mass balance, energy balance, capital cost and operating cost of an industrial-scale HTC plant. The process model was developed with flexibility in mind, capable of working with a wide range of custom feedstocks, HTC process conditions and plant sizes. This model was validated using industrial cost and energy use data, offering large improvements in accuracy over the few currently existing process models for HTC. Through researching the HTC product markets and feedstock costs or savings, investment analyses were performed to show the potential profitability of an industrial-scale HTC plant.

HTC is particularly appropriate for wet wastes and can offer significant savings compared to current treatment and disposal practices. Digestate, the by-product of the anaerobic digestion (AD) process, is generated in increasing quantities in the UK, and there are growing concerns over the costs and restrictions on its use and management. Therefore, it was selected as an interesting HTC feedstock to focus on and explore further through a detailed case-study. From this, recommendations and thresholds were determined for the treatment of digestate by HTC in the UK, also giving an indication of the potential for HTC on the whole.

1.2 Hydrothermal carbonisation

Hydrothermal carbonisation is a thermochemical process that converts wet, waste biomass into a solid hydrochar, process liquid and a small amount of CO_2-rich process gas (Figure 1.1). Hydrochar has similar properties to lignite coal [3] containing 45–75% of the carbon initially present in feedstock [4]. The process liquid is a mixture of mostly sugars and organic acids [5]. The product fractions and properties are governed by the feedstock used and HTC process conditions. Hydrochar has potential markets as a solid fuel, soil amendment, fertiliser and activated carbon. The process liquid can be treated and disposed of, recirculated into the HTC process or marketed as a fertiliser or a substrate for AD. To date, no use has been found for the process gas consisting of >95 wt.% CO_2, and this is typically vented to atmosphere.

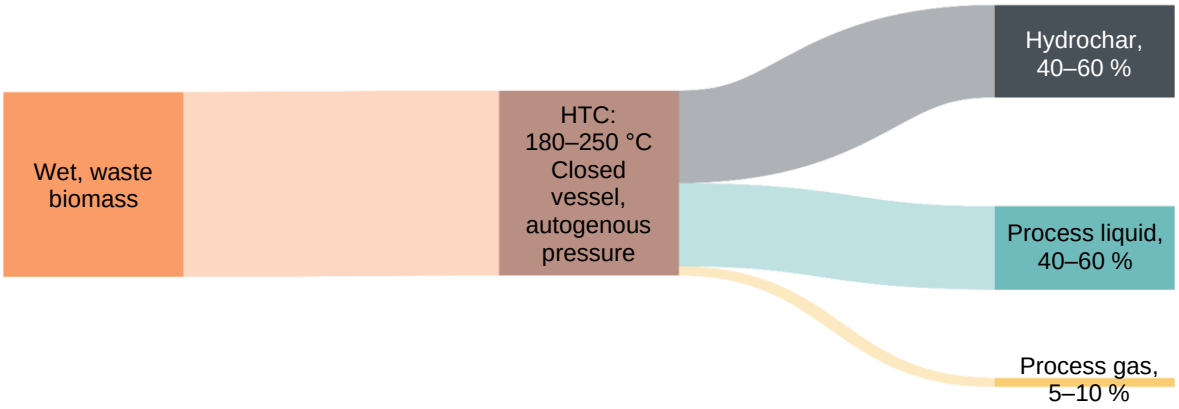


Figure 1.1: The HTC process with approximate product fractions.

1.3 Wet waste generation in the United Kingdom

Recycling rates in the UK continue to increase with yearly reductions in household waste sent to landfill. However, the generation of waste itself has remained relatively constant over the past 10 years [6]. Rather than only considering landfill or recycling, there has been a recent focus on the exploitation of waste with the view for creating useful products [7]. Biodegradable wastes, in particular, have been identified as a commercial opportunity by the House of Lords due to their valorisation potential [8]. Figure 1.2 shows the trend in some of the main fractions of biodegradable waste in the UK between 2010 and 2016. It was assumed that household waste consisted of 65% biodegradable content [9,10]. Large quantities of biodegradable waste are generated in the UK and have remained fairly consistent over recent years. This is a sizable and stable market for emerging technologies such as HTC, capable of forming high-value products. Case-studies for specific types of biodegradable waste will help to better understand the market and explore the potential economic benefits from using HTC or similar technologies.

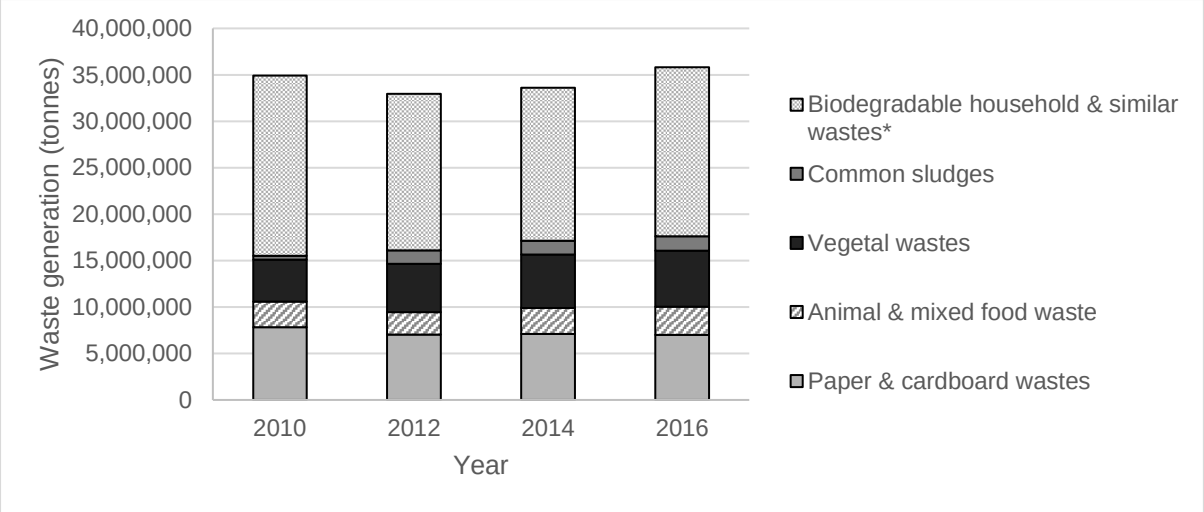


Figure 1.2: Common streams of wet waste generation in the UK between the years 2010 and 2016 [6]*. **assuming 65% of the total household & similar wastes are biodegradable.*

1.4 Aims and objectives

The overall aim of this research is to investigate the technical and economic feasibility of HTC for the treatment of wet, waste biomass, with a particular focus on digestate from AD as a feedstock. This aim was achieved with the development of a fully flexible, techno-economic model, based on experimental HTC data and validated by industrial cost and energy use data. The objectives of this research were to:

- Develop a clear experimental methodology to investigate the HTC of three types of digestate at different temperatures, residence times and moisture contents.
- Measure the product characteristics, with a particular focus on hydrochar and process liquid markets.
- Investigate the dependence of product characteristics on the HTC process conditions for each of the types of digestate through the use of regression models.
- Build a predictive process model based on the regression models and publicly available sizing and cost data.
- Validate the process model using industrial mass balances, energy usages and cost data.
- Identify and estimate the sale price of products across various markets.
- Assess the current status of AD and digestate production across the UK.
- Perform an investment analysis for the HTC of digestate in the UK, based on the expected product sale prices and savings available for diverting digestate from current management and disposal methods.
- Consider the effect of plant scale, economies of scale and the cost of transporting the digestate to the HTC plant.

1.5 Thesis overview

This thesis is split into different chapters covering: the current literature, a clear methodology behind the work, the results of the experimental, process modelling and investment analysis, and a case-study considering the HTC of digestate. These chapters are followed by a conclusion covering the main findings and areas for future work and finally an appendix with detailed experimental results. More details on each of the chapters is given below.

Chapter 2 Literature Review

This chapter gives the background and critically reviews literature on the following topics:

- wet waste generation in the UK,
- the HTC process and the reactions involved, including the effect of feedstock type and process conditions on the HTC process and its products,
- various methods and models that have been developed to predict these effects,
- HTC products and their uses, including desirable product characteristics for each market,
- the techno-economics of HTC, including existing mass balance, energy balance and economic models.

The literature review introduces the topics considered throughout the rest of the thesis, as well as highlighting the gaps in current understanding that are covered in this work.

Chapter 3 Methodology

The methodology covers the steps taken in the experimental, process modelling and economic assessment work. It details the three types of digestate feedstock used experimentally as well as the rationale behind the HTC process conditions. The procedures for HTC, product characterisation and germination and plant trials are all detailed. Each of the unit operations included in the process model are listed along with their sizing calculations and capital cost estimation tools. The UK standards used for approximating operating costs are given. A consideration for the markets of both the products and feedstocks is also reported.

Chapter 4 HTC Test Programme

This chapter details the experimental results. There is a focus on the mass yield and carbon content of the hydrochars produced, as this is a good indicator of its quality and value. The fate of both organic and inorganic elements is also measured. The suitability of hydrochar and process liquid in a variety of markets is assessed, with a particular focus on elemental

concentrations and results of the plant trials. The regression models developed from the HTC experiments are presented, as well as a comparison with models in literature.

Chapter 5 HTC Plant Model

The process model is based on the regression models from experimental results and publicly available data on plant design and costing. This chapter covers the development of the plant model and the results it gives for performance, capital costs and operating costs, initially for default plant conditions. It is validated using industrial data and compared to literature results. Then, the optimisation of the process model is explored, considering the product properties, plant costs and plant capacity. As the plant model was designed with flexibility in mind, the performance and costs are also reported for alternative feedstocks, different to the digestate feedstock results the model was built upon.

Chapter 6 Case Study: Feasibility of HTC as a treatment for AD digestate

AD digestate has been identified as a promising feedstock for use with HTC. The status of digestate generation in the UK is assessed, including the processing costs currently incurred during digestate management. The dewatering of digestate is also discussed, considering its importance prior to HTC and the costs that are involved. The cost of transporting digestate to a HTC plant is estimated, using Lincolnshire and Nottinghamshire as examples. The digestate market findings are incorporated into an investment analysis. The payback periods under a range of plant and market conditions are reported. There is a particular focus on the impact of the product and feedstock costs on the economic feasibility of the HTC of digestate, as well as a consideration of other feedstocks. From this, the range of conditions necessary to achieve a defined payback period are reported. This is very useful for understanding the economic feasibility of HTC and its future potential in the UK market.

2 Literature Review

2.1 Introduction

This chapter reviews the current status of waste production in the UK and the literature surrounding the hydrothermal carbonisation process, its products and techno-economic viability. An overview is given of:

- The availability of waste in the UK, with predictions of its potential value.
- The HTC process, including the proposed reaction pathways and differences compared to other thermal treatments.
- Models developed to explain the effect of varying feedstock characteristics and changing process conditions on HTC products.
- The target properties of hydrochar destined for use as a solid fuel or for soil application.
- The properties of process liquid, the effect of recirculation in HTC, and its application in anaerobic digestion.
- Existing techno-economic models of HTC.

While the understanding of HTC has increased rapidly over the years, there is still the need for a deeper understanding of HTC products in various applications and a validated techno-economic model, capable of predicting its potential in a range of scenarios.

2.2 Availability of wet biomass waste

The Waste Framework Directive 2008/98/EC defines waste as “any substance or object which the holder discards or intends or is required to discard”, and its generation across the globe is rapidly increasing [11]. Alongside calls for reduction in waste generation, there is an increased interest in the exploitation of waste, not only for the creation of useful products, but also as a way of negating the environmental concerns of landfill [7]. While the recycling and reuse rate has generally increased in the UK, 14.6 million tonnes of municipal waste was sent to landfill, including 7.2 million tonnes of Biodegradable Municipal Waste (BMW) in 2018 [6]. This is a 20% of the baseline 1995 value, meaning the country is on target for the EU goal as set out in the Landfill Directive 1999/31/EC [12], but it is still higher than many other European countries. Indeed, this large amount of BMW has been identified as a commercial opportunity by the House of Lords due to its valorisation potential [8].

There are several potential uses for bio-waste, including: direct energy production (heat and/or electricity); fuels (e.g. biogas, bioethanol); chemicals (speciality or bulk); and others (animal feed, fibres, soil amenders etc.). These feedstocks have been valued at $60–150/tonne biomass for electricity generation, and up to $1000/tonne biomass for bulk chemicals

production [7]. Defra estimated that of the 100–150 million tonnes of carbon-containing waste generated in the UK in 2010, 100 million tonnes could have been used for biogas production [8]. There are already growing markets for the valorisation of agricultural waste, but the commercial development of municipal solid waste (MSW) and sewage sludge treatments is slower due to the heterogeneous nature and high moisture content of the materials. However, with a growing population and increasing urbanisation across the planet, generation of these waste resources is set to increase substantially. This is particularly true for economically developing countries whose waste contains more organic matter than in other parts of the world [7].

The valorisation of waste is not merely a scientific or industrial matter, but also a policy priority. The House of Lords has expressed the need to perceive waste as a resource and to calculate its true worth so it can be valorised in the most advantageous ways, rather than just the simplest. In a report focussing on the bio-economy, they concluded that there are substantial environmental and economic benefits in exploiting carbon-containing waste, with the industry having an estimated worth of £100 billion [8]. They conducted work with the Department for Business, Innovation & Skills on how to build a high value bioeconomy [13]. This highlighted technologies such as Anaerobic Digestion (AD) for the treatment of wet waste. AD is a technically mature process, with proven benefits over some other waste treatments [14]. It is capable of treating a wide variety of wet wastes, avoiding the costly and energy-intensive drying step prior to incineration. However, it results in the generation of large quantities of digestate as a by-product. In 2019, 12.5 million tonnes of feedstock were processed by 486 AD plants resulting in the generation of over 500,000 dry tonnes of digestate [15,16] , containing between 130,000 and 240,000 tonnes of carbon [17,18]. As of April 2020 the number of plants reached 579 with a further 331 under development [19]. Currently, the majority of digestate produced is transported and spread to land, coming in at a cost of £16/tonne when transported an average round-trip distance of 50 miles [20,21]. There are restrictions to spreading and storage requirements in addition to this which incur further costs [22,23]. A large number of commercial and industrial AD operators have their digestate removed by off-site companies at a cost [24]. AD capacity in the UK has increased over the last 15 years, supported by feed-in tariffs and renewable heat incentives [15]. It is expected to increase further over the coming years, especially for the treatment of food waste [9]. Food waste AD plants tend to be large-scale and located close to population centres, producing larger quantities of digestate than can be spread in the local area. Nonetheless, the House of Lords has suggested that caution be taken with AD technology, as for many types of waste there are potentially better treatments that could produce higher-value products. They warned the government about distorting the market and creating a situation similar to that in Germany

where the AD plants are so heavily subsidised that the country has started to grow crops specifically as feedstock for AD installations [8].

There is therefore interest in alternative low-cost biomass treatment processes suitable for wet wastes. HTC has been highlighted as a promising and energy-efficient treatment for wet wastes and the production of chars [25]. With an abundance of wet wastes generated in the UK, it is a technology deserving of high interest.

2.3 The HTC process

HTC was first described in 1913 by Friedrich Bergius, a German chemist and recipient of the Nobel Prize in Chemistry (1931) for his work on high-pressure methods [26]. Commercial interest in HTC can be dated back to at least 2008 with the emergence of patents relating to the technology [27–29]. The early patents were filed by inventors based in Germany, still very much a global hotspot for industrial HTC technology today. However, it has not been until the past 15 years that the reaction mechanisms, kinetics and product qualities have been investigated. Figure 2.1 shows the steep rise in journal articles containing the term HTC.

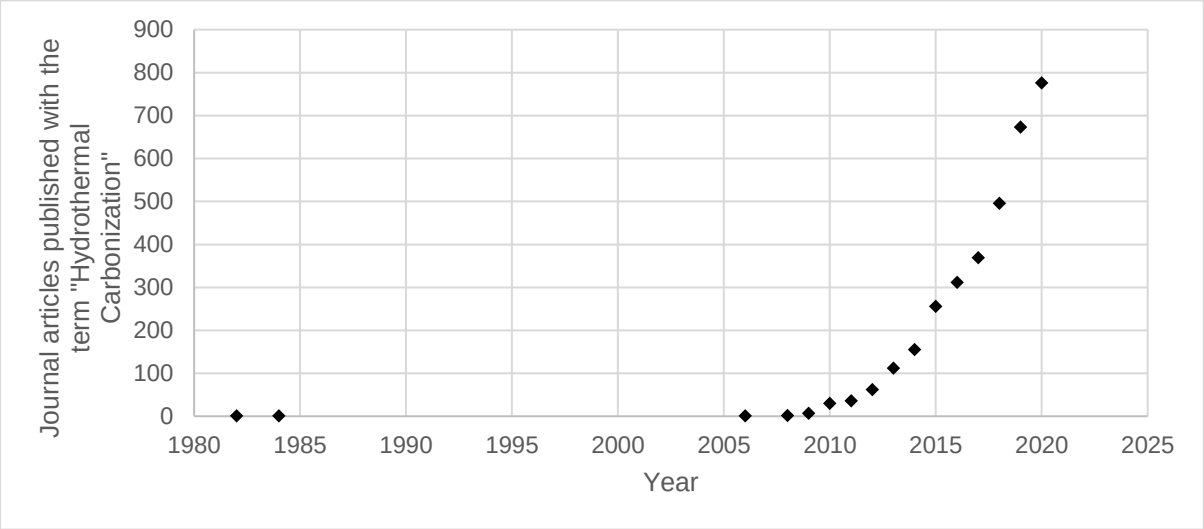


Figure 2.1: Number of journal articles containing the term "hydrothermal carbonization/carbonisation" according to records on ScienceDirect.

2.3.1 Reactions

The main stages of HTC are hydrolysis, dehydration, decarboxylation, aromatisation, and condensation-polymerisation. The nature and relative significance of these mechanisms depend on the feedstock used [30]. Hydrochar is hydrophobic in nature and has proven stability in water [31]. Along with hydrochar, there is evidence of a secondary char formation, especially at higher temperatures and moisture contents [21]. Sevilla and Fuertes [32] conducted one of the pioneering studies on HTC, specifically the HTC of pure cellulose. By

testing at different temperatures and observing the hydrochar, they deduced that the onset of hydrothermal reactions for cellulose occurred between 210 °C and 220 °C at autogenous pressure. They observed that the degree of carbonisation, i.e., the carbon content of the hydrochar, increased with temperature. Over the whole temperature range, the hydrochar mass yield was 30–50 wt.%, and the carbon yield was 54–84 wt.%. Further work on the HTC of lignocellulosic biomass has concluded that hemicellulose degrades at around 180 °C and lignin at around 200 °C [30]. Lei et al. [33] proposed the possible reaction pathways of HTC, based on their investigation of corn stalk, depicted in Figure 2.2. This suggests that the hydrochar solid product consists of two forms of char. The polymerised hydrochar was created through a series of aqueous reactions, but the porous polyaromatic char was formed from non-dissolved cellulose and lignin via a heterogeneous pyrolysis-like process. HTC is exothermic in both pure compounds and municipal waste streams [34,35]. HTC is of growing interest and the conversion of many wet feedstocks has been studied to produce a number of materials from coal-substitutes, to soil amendments and carbonaceous materials for chemical applications [5].

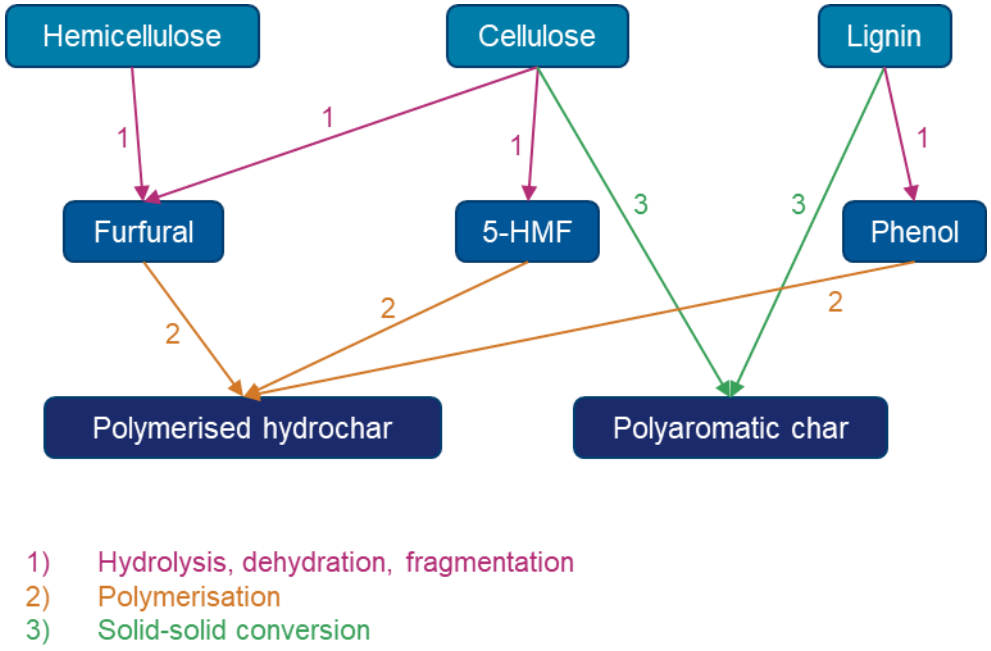


Figure 2.2: Possible reaction pathways to form hydrochar, based on Scheme 2 from Lei et al. [33]

The primary difference between HTC and other thermochemical processes is that it is performed in an aqueous environment. HTC is more energetically favourable for wet feedstocks compared to pyrolysis, gasification and other dry thermochemical processes [25]. Pyrolysis and gasification are more established biomass treatments than HTC, producing high

value chemicals. However, a costly drying process would be required as a precursor for wet feedstocks. Table 2.1 highlights the key differences between HTC, pyrolysis and gasification.

Table 2.1: Comparison of pyrolysis, gasification and hydrothermal carbonisation.

	Pyrolysis	**Gasification**	**HTC**
Conditions	Dry	Dry	Wet
Temp.	400–900 °C [14]	>750 °C	Typically 180–250 °C [30]
Products	Bio-oil: (29–38 MJ/kg) [36] Biochar: used as adsorbent or adsorbent precursor, soil conditioner or carbon sequestration [37] Py-gas: CO_2 (40–60%), CO (15–35%), H_2 (5–25%) and CH_4 (5–10%) [38]	Syngas: primarily CO, H_2 and CO_2 along with significant quantities of CH_4, N_2, O_2 and H_2O (4–6 MJ/m^3) [39] Low-carbon ash by-product	Hydrochar: similar properties to lignite [3] containing 45–75% of the carbon initially present in feedstock [4] Process liquid: mostly sugars and organic acids [5] CO_2-rich gas
Notes	Can target bio-oil (fast pyrolysis) or biochar (slow pyrolysis) production [14]	Syngas composition and energy content heavily depend on the feedstock used and gasification conditions Syngas often must be cleaned of trace-level impurities prior to downstream gasification processes [39]	Carried out in a sealed vessel at autogenous pressure. Subcritical water acts a catalyst [5] Weak acids (e.g. citric acid) can act as a catalyst [40] Usually takes several minutes to several hours [3]

2.3.2 Feedstock type and process conditions

Experimental HTC studies have spanned different types of wet biomasses, including pure compounds such as cellulose [32], other lignocellulosic materials [41], municipal wastes [34], sludges [42] and digestates [43]. Many of these have investigated the effect of varying process conditions on the reaction products. The desired product characteristics will very much depend on the target market, but generally hydrochar with a higher carbon content is of greater value. At the same time, hydrochar yield is also an important factor. There is unanimous agreement that temperature is the most influential factor on the reaction products [5,41–53]. Generally, it has been found that an increase in temperature leads to an increase in the degree of carbonisation, resulting in hydrochar with a higher carbon and energy content. However, the mass yield of hydrochar decreased at higher HTC temperatures, presenting an important

trade-off in hydrochar quality and quantity. While temperature is almost always considered in literature, other commonly studied factors are the residence time, moisture content of feedstock and the pH. Findings relating to these factors are more mixed. Some studies found the feedstock moisture content to be significant with an increase in moisture leading to an increase in the degree of carbonisation [54,55], another found moisture content to only be significant at certain values of temperature and time [52], and still others concluded that it was insignificant [44,47,50]. Most researchers found an increase in time to have an effect much like increasing the temperature, although to a lesser degree: a decrease in solid yield but increase in carbonisation [42,44,45,47,50,52]. It was found that a lower pH resulted in a higher degree of carbonisation and an increase in the removal of ash from the hydrochar [56,57]. The effect of pH on hydrochar yield was dependent on temperature, with a lower pH at higher temperatures increasing the yield but decreasing the yield for temperatures <200 °C.

While many of these findings are based on simple observations of the results, some researchers have investigated the effect of process conditions in greater depth by generating kinetic, statistical or computational models. The process conditions of HTC have a large effect on the product properties, but significant variations are found between the different feedstocks. In order to predict the mass balance and product properties of HTC at a range of conditions, there are two main approaches: a detailed feedstock-specific model, or a universal model designed for all feedstocks.

Many of these models are based on regression analyses. Multiple linear regression is an explicit technique that results in the generation of an easy to use linear equation for dependent and independent variables. However, a major drawback of this approach is that it assumes a simple linear relationship between variables which can lead to significant errors in the results. The general form of equations is shown in Eq. 2.1. By using enough experimental data points, interaction effects can be included in the model. Box-Behnken and Central Composite Design are two different designs of experiments that are commonly used to reduce the number of runs required to sufficiently investigate the different factors.

$$Y_i = \alpha + \beta_1 X_{i,1} + \cdots \beta_p X_{i,p} \qquad \textit{Eq. 2.1}$$

where α is a constant, β is the coefficient and X is the factor of which there are i number.

Regression trees are binary trees that split dependent variables into nodes. This has an advantage in that no prior relationships between variables are assumed, unlike with multiple linear regression, and so nonlinearities can be modelled. However, they are discontinuous, give discrete results, and can be more complex to use. Random forest models are tree-based models that consist of a large collection of regression trees. They have been described as a

robust black box approach [58]. They are a more comprehensive version of a regression tree but have the same issues with discontinuity and discrete results.

2.3.2.1 Feedstock-specific models

Typical biomass-specific models are based on the principle of a severity factor (SF). The severity factor is usually defined as the logarithm of the reaction ordinate, R_0, a term defined as a means of weighting the time and reaction temperature by an arbitrary rate process. The most commonly used reaction ordinate is that developed by Overend and Chornet in 1987 [59], characterising the fractionation of lignocellulosic biomass by steam-aqueous pre-treatments based on the Arrhenius relation (Eq. 2.2).

$$R_0 = t \times \exp\left[\frac{T - 100}{14.75}\right] \quad \textit{Eq. 2.2}$$

$$\mathrm{SF} = \log R_0 \quad \textit{Eq. 2.3}$$

where R_0 is the reaction ordinate, t is the retention time (minutes), T is the reaction temperature (°C) and SF is the severity factor.

The severity factor works on the basis of different process conditions having non-linear effects, specifically an exponential effect for temperature. This severity factor was used by Hoekman et al. [31] when studying the HTC of loblolly pine. They found that when comparing the energy content of hydrochar from 6 different sets of process conditions, there was a common relationship with the calculated severity factor. Jeder et al. [60] also used this severity factor when assessing the effect of temperature and time on the HTC of olive stone. They found linear relationships for solid yield and O/C ratio with respect to severity factor.

Suwelack et al. [40] used an adapted form of the severity factor to develop a regression equation for the HTC of anaerobic digestate. They worked on the basis of the more complex equation developed by Abatzoglou [61] (Eq. 2.4), based on the Taylor series expansion, that also considered acidic and catalytic parameters, assuming isothermal conditions. They varied temperature, time and used citric acid as a catalyst, setting concentrations to result in a pH of 3, 5 and 7.

$$R_0 = \exp\left[\frac{X - X_{ref}}{\lambda X_{ref}}\right] \times \exp\left[\frac{T_p - T_{p,ref}}{\omega}\right] \times d_R \quad \textit{Eq. 2.4}$$

where X is the catalyst concentration, T_p is the reaction temperature and d_R is the residence time. Reference values were defined by Suwelack [40] as 0.548 g citric acid per g digestate (equivalent to a pH of 3) and 190 °C.

They found temperature to be the dominant factor for hydrochar mass yield and degree of carbonisation. Citric acid concentration had the second largest impact on mass yields followed by residence time. They called for further studies on other feedstocks and also investigations into the impact of particle size, moisture content and presence of ionic salts.

One of the major criticisms of these severity factor-based equations is that, in effect, temperature and time can act as exchangeable parameters [30]. While temperature has an exponential effect on severity factor, the equation could be exploited by increasing the retention time enough to account for an elevated temperature. This is problematic as certain specific temperatures are required for the different chemical changes necessary for HTC [30].

Mumme et al. [43] investigated the effect of reaction temperature, residence time and pH on the hydrochar yield and carbon content with the HTC of maize silage digestate. A Box-Behnken design of experiments was used along with a p-value of 0.05 to measure the significance of each factor. All terms were deemed significant other than the pH for the hydrochar carbon content. The developed equations had R^2 values of 0.98 and 0.97 respectively, indicating that they gave a good estimation of parameters. They had standard errors of 2.01% 1.74% for the hydrochar yield and carbon content, respectively. When validating the model with a further three HTC runs, they found these had a mean deviation of 2.81% and 1.54% respectively. As these were comparable with the models' standard deviations; the accuracy of the regression model was deemed proven by the researchers.

Volpe et al. [62] performed a multivariate linear regression for the mass yield and energy content of the hydrochar, and the energy yield, $mass\ yield \times \frac{HHV_{hydrochar}}{HHV_{biomass}}$. They used temperature, time and feedstock moisture content as factors. Regression results (based on a p-value of 0.05) showed that all three factors were significant for estimating the hydrochar yield, temperature and time were significant for energy content, and only the moisture content was significant for the energy yield.

Toptas Tag et al. [44] considered the role of temperature and time on the mass yield and energy densification ratio, $\frac{HHV_{hydrochar}}{HHV_{biomass}}$, of three types of biomass: sunflower stalk, poultry litter and algae. They used a Central Composite design of experiments. The produced quadratic models showed that temperature was always a significant factor across all the feedstocks, time was often significant, moisture content was never significant, and interaction effects including temperature and time were occasionally significant.

Each of the studies discussed up to this point have been dimensionless, assuming a constant reaction temperature throughout the experiment. But in reality, there will be variations in

temperature within the reactor, and this variation is likely to influence the localised behaviour of the HTC process. The type of reactor, reactor size, method of stirring and heating rate will all affect the distribution of temperature. Álvarez-Murillo et al. [63] created a three-dimensional computational model of the HTC of cellulose. The model fitted the experimental data well and temperature was, again, found to be the most important process condition. The results also confirmed that there is an induction period in heating up the reactor, and they suggested that this heating up time should be taken into consideration, rather than just simply modelling with a constant temperature period assumption.

The feedstock-specific studies detailed here have generated relationships and regression models with often a very good fit and low error. They can be used to build up a general picture of how HTC process conditions affect the product properties but a HTC operator could not use the model results and apply them directly to different feedstocks. The generation of a universal model that could be applied to a range of feedstocks would be much more useful for researchers and operators investigating different feedstocks and HTC process conditions.

2.3.2.2 Universal models

Both Heidari et al. [64] and Guo et al. [54] used the severity factor (Eq. 2.2) to create models for a range of feedstocks. Heidari [64] used artificial mixes of varying concentrations of cellulose, hemicellulose and lignin to represent real biomass samples. They found the biomass composition to have a larger effect than severity factor on the mass yield, carbon content and HHV of the hydrochar. While their regression models serve to understand the details of the HTC of pure lignocellulosic compounds, the high ash content typically found in many wet waste biomasses was not considered and therefore the application of their model is limited. Guo et al. used the dose-response method with different lignocellulosic feedstocks, modelling the hydrochar yield, carbon content and HHV. Their models gave a good representation of the feedstocks studied, but as the dope-response method does not result in a regression equation, it is still difficult for other researchers to apply the findings.

Li et al. [65] aimed to create universal statistical models in which the feedstock properties and process conditions were used to determine the yields and energy content of the products. They gathered data from 313 literature studies and analysed these using multiple linear regression and regression trees. They used data from both lignocellulosic and non-lignocellulosic feedstocks to generate their models. Regression models were generated for solid hydrochar yield, hydrochar carbon content, hydrochar energy content and the normalised carbon contents of the hydrochar, process liquid and process gas. The R^2 and root mean squared error values were calculated for each of these. R^2 values were higher and root mean

squared error values were lower for the regression tree models, indicating that these gave better results. They concluded that both types of regression model fitted the data well. While the models could be used as general guides or screening tools, their use beyond that is somewhat limited. The range of factors was extensive, including those not commonly reported such as reactor heating rate. If an operator wanted to make predictions using the multiple linear regression, they could omit these terms from the equation on the assumption that the effect was minimal. However, for the regression trees, each parameter must be known in order to follow the binary splits correctly. Also, the inability to fine-tune is apparent with the regression tree. If a user wanted to investigate the effect of a change in temperature for the same feedstock, the regression trees would often give the same discrete result.

Li et al. [58] went on to develop their work further, producing improved linear and non-linear models for predicting hydrochar properties from process conditions and feedstock properties. They updated their database of experimental data from literate and this time recorded the chemical composition (cellulose, hemicellulose and lignin content) of the feedstocks along with the ultimate and proximate properties. The feedstock polarity index (Eq. 2.5), as defined by Rutherford et al. [66], was approximated from the collected data. The smaller the polarity index, the more hydrophobic the feedstock.

$$Polarity\ index = \frac{Polar\ groups}{Apolar\ groups} = \frac{O + N(+S)}{C} \qquad \textit{Eq. 2.5}$$

where O, N and C represent the weight percentage of oxygen, nitrogen and carbon of the feedstock respectively. Sulphur, S, is sometimes included in the equation.

The hydrochar yield, carbon content and energy content were modelled. In this study, they again used multiple linear regression models to describe linear relationships, omitting correlated parameters. Non-linear relationships were approximated using regression trees and random forest models. The models were evaluated using the R^2 value, mean absolute percentage error and root mean squared error. Sobol analysis, a Monte Carlo-based variance decomposition method, was used to identify the parameters with the greatest influence on the results. The forest models gave the best results in terms of a higher model fit and lower root mean squared error. This supports the theory that the relationship between char yield and feedstock properties and process conditions is non-linear. The Sobol analysis results suggested that hydrochar yield was most sensitive to initial solids concentration and reaction temperature. The hydrochar carbon content was highly sensitive to feedstock carbon content (accounting for 50–70% of the variance), ash content and reaction temperature. For the hydrochar energy content, the feedstock and hydrochar carbon contents were found to be the

most influential parameters. This study was conducted with greater depth, producing more robust results. However, aside from the useful results on the relative influence of parameters, there is little else to be taken by the reader as the regression models themselves are not reported.

Vallejo et al. [67] presented another study using a multiple linear regression model and regression tree. The models could be tailored for both lignocellulosic and non-lignocellulosic feedstocks through the inclusion or omission of the reactivity index (Eq. 2.6). Additionally, they included the sulphur content in the equation for polarity index, omitted by Li et al. [58]. They calculated the reaction ordinate, R_0, as defined by Belkacemi et al. [68] and Leger et al. [69] (Eq. 2.7). The severity factor (SF) was taken as the logarithm of this (Eq. 2.3).

$$Reactivity\ index = \frac{Reactive\ part}{Inert\ part} = \frac{H_0 + S_0}{L_0 + A_0} \qquad \textit{Eq. 2.6}$$

where H_0 is the hemicellulose, S_0 the aqueous extractives, L_0 the lignin and A_0 the ash content of the raw biomass.

$$R_0 = t \times \exp\left(\frac{T - T_{ref}}{\omega}\right) \qquad \textit{Eq. 2.7}$$

where t is the temperature, T is the reaction time, T_{ref} the reference temperature and ω the temperature effect constant.

The models were created to predict the hydrochar yield and energy content from the feedstock properties and process parameters. Similarly to Li et al. [65], the fit of the model was evaluated using the R^2 value along with the mean absolute percentage and root mean square errors. The model was created using 70% of the collected data and validated with the remaining 30% of the data. Models were developed for non-lignocellulosic materials without including the reactivity index. In general, the regression trees gave better results than the multiple linear regression models when considering the R^2 and root mean square error. Additionally, the model gave a better fit for lignocellulosic biomass, including the reactivity index, rather than non-lignocellulosic biomass which omitted this.

The results of these universal models agree with the findings of feedstock specific models: an increase in process severity led to a higher degree of carbonisation but a lower hydrochar yield. A comparison of the relative weighting between process conditions and feedstock properties for Li et al. [65] and Vallejo et al. [67] is shown in Figure 2.3. These weightings are taken from the regression trees produced in both models and show a general agreement:

process conditions are more influential for mass yield but feedstock properties are more influential for hydrochar HHV.

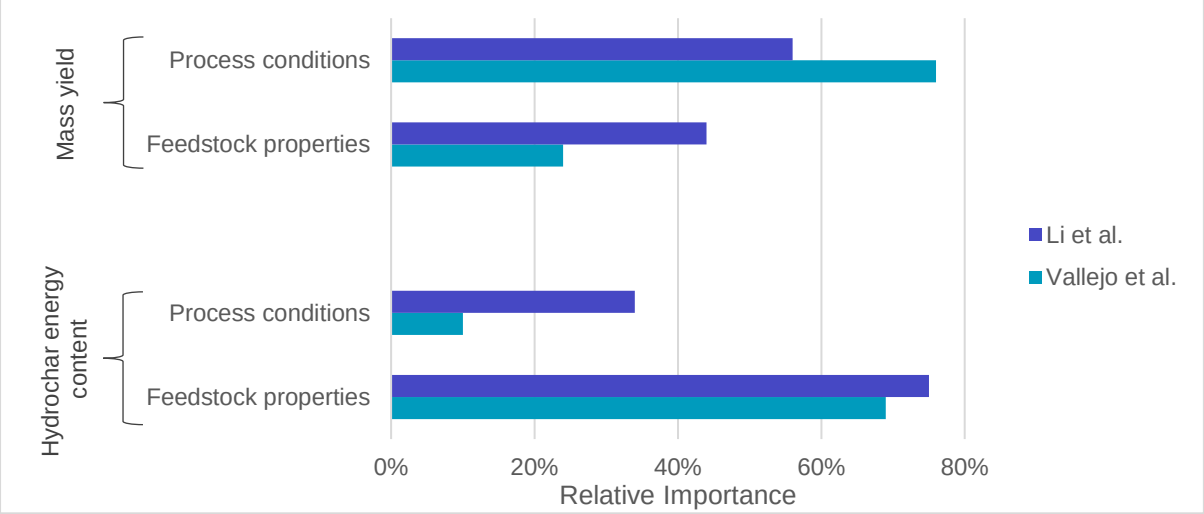


Figure 2.3: Relative weightings of process conditions for the mass yield and hydrochar HHV found by Li et al. [65] *and Vallejo et al.* [67].

These universal models are useful in that they can be used to predict product properties and yields, and give a good indication of the relative weighting of feedstock characteristics and process conditions for a whole range of feedstocks. However, their wide applicability comes at the detriment of accuracy. Feedstock-specific models developed using experimental results tend to give much more accurate results for the relevant feedstock. However, their development is more laborious and time-consuming. Ultimately, there is a trade-off between the higher accuracy of feedstock-specific models and the simplicity of universal models. In practice, it could be that a universal model be used as a screening tool for multiple feedstocks, and then if better predictions for a particular feedstock were desired, a feedstock-specific model could be developed using the experimental and statistical methods discussed in Chapter 3 Methodology Section 3.3.

2.4 HTC products and their uses

HTC results in two primary products: solid hydrochar and process liquid. As discussed, altering the feedstock used and/or process conditions affects the quantities and properties of these products. The optimum operating conditions will depend on the feedstock used and the target market of HTC products. This section describes the main uses of hydrochar and process liquid from wet waste biomass, covering the desired characteristics and suitable operating conditions.

2.4.1 Hydrochar combustion

The combustion of hydrochar as a solid fuel is the most established and researched product stream to date. When using biomass fuels in pulverised fuel boilers, properties similar to coal are desired, particularly high energy content, low water-retention, low reactivity and low volatile matter content [70]. One of the largest barriers to the combustion of biomass materials is the high alkali and alkali earth metals concentration in the ash, proven to cause slagging and fouling issues in the boiler [71]. In order to reduce the issues associated with slagging, a high ash melting temperature is desired. The alkali and alkali earth metals have a large influence on ash melting temperature and it has been shown that generally, potassium and sodium reduce it, while magnesium and calcium increase it [72]. Fouling is brought about when potassium and sodium combine with chlorine to form alkali chlorides, which condense on heat exchanger surfaces causing corrosion. Therefore, a reduction in potassium concentration of the hydrochar theoretically reduces the risk of fouling also [73]. HTC is one potential method of improving the fuel characteristics of biomass materials.

Smith et al. [73] studied the fate of inorganic material during the HTC of both lignocellulosic (miscanthus, willow and oak wood) and non-lignocellulosic (food waste, sewage sludge, AD press cake, algae and greenhouse waste) feedstocks. The hydrochars of each of these samples had higher energy contents but also higher ash contents than the starting material. The difference in results at 200 °C and 250 °C depended on the feedstock in question. The hydrochars from the plant-based feedstocks had HHV values of 25–32 MJ/kg and ash contents of <3.5%. The HHV of the AD press cake and sewage sludge hydrochars reached a maximum of 9 MJ/kg and ash contents of 72–75% and 19–34% respectively. They observed a significant removal of alkali metals during HTC. However, a greater proportion of organic material was removed from the hydrochar than inorganic material, thus increasing the overall concentration of ash. They observed a significant extraction of potassium and sodium from the feedstock, but magnesium, calcium and phosphorous were removed in more limited amounts. Due to the reductions in these metals, the hydrochars were shown to have higher ash melting temperatures than the original feedstocks, reducing the propensity to slagging.

Stirling et al. [70] investigated the char reactivity of hydrochars from the HTC of wood pellets and olive cake feedstocks at 200 °C and 225 °C. The HTC of wood at 225 °C led to the largest metal extraction of 89% for sodium, 93% for magnesium, 97% for potassium, and 92% for calcium, with the majority of these elements ending up in the process liquid phase. They found that the hydrochar burned over a longer time than the original feedstock, with the wood hydrochar burnout being comparable to bituminous coal. The char reactivity was measured using thermal gravimetric analysis (TGA) at low heating rates and a drop-tube furnace (DTF)

at much higher heating rates, with similar conditions to a pulverised fuel boiler. Hydrochar produced at 225 °C had an almost identical reactivity to that of bituminous coal. This was primarily caused by the removal of catalytic alkali and alkaline earth metals. While the reactivity of the hydrochar matched that of the coal, further torrefaction was required to reduce the volatile matter of the hydrochar samples. The torrefied hydrochar samples resulted in volatile matter contents between that of sub bituminous and high-volatile bituminous coal, a reactivity similar to that of high-volatile bituminous coal and a low ash content, at <0.5% compared to that of 5.2% for bituminous coal. The torrefied hydrochar samples had better solid fuel properties than the original feedstock.

Smith et al. [74] investigated the production of high-quality hydrochar from miscanthus. The miscanthus feedstock was diluted to a moisture content of 90 wt.% from the as-received moisture content of 33.6–57.7 wt.%. They found that hydrochar produced at 250 °C had a HHV of up to 28 MJ/kg. There was a significant reduction in alkali metal content in the hydrochar compared to the raw biomass, increasing the safe combustion temperature and reducing the theoretical risk of slagging, fouling and corrosion. Miscanthus is typically harvested in late winter/early spring for fuel applications to ensure a lower ash content of the crop. Using the HTC process opened up the option of early harvesting of miscanthus, bypassing the overwinter dry matter losses and allowing for increased biomass yields.

While these studies have focussed generally on low-ash feedstocks, the combustion of hydrochar from high-ash materials has also been investigated. Both He at al. [75] and Zhuang et al. [76] used sewage sludge with a high moisture content of 85.7 wt.% and 90.9 wt./% respectively in HTC to investigate the production of a clean fuel. Through the analysis of weight loss and weight loss rate curves, they concluded that the combustion of hydrochar was more stable than the original sewage sludge feedstock due to the reduction in volatile matter. He et al. [75] found that a residence time of 6–8 hours gave the best performance in terms of combustion performance at 200 °C. Zhuang et al. [76] studied the impact of both temperature and time on the produced hydrochar. They found that the combustibility index, a factor of mass loss rates and ignition and burnout temperatures, was insufficiently high to be considered as a good quality fuel product. However, HTC temperatures of lower than 240 °C were highlighted as the most promising. This shows the trade-off between high energy content at higher HTC temperatures and the other beneficial combustion characteristics that must be considered.

The combustion of high-ash hydrochar from the HTC of municipal sewage sludge was proposed by Wang et al. [77]. 230 °C was determined as the optimal HTC temperature to produce hydrochar for combustion according to the fuel characteristics (relatively high energy content, low calcium and magnesium content, lower volatile matter, higher fixed carbon

content, and improved dewaterability) and potential for AD of the process liquid. They performed HTC on mechanically dewatered sewage sludge with an ash content of 55 wt.% on a dry basis. The HHV of the feedstock on a dry basis was 6.3 MJ/kg. The relative ash content of the hydrochar was higher than that of the feedstock, from 58 wt.% at 170 °C up to 69 wt.% at 350 °C, but the absolute ash content had reduced due to some ash dissolving into the process liquid, especially the alkali and alkaline earth metals. The calcium and magnesium content of the hydrochars decreased to a minimum at 230 °C and increased at higher temperatures. There was also significant removal of sodium and magnesium containing compounds, reducing the propensity of the hydrochar to slagging and fouling. The HHV of the hydrochar reached a maximum at 230 °C at 5.5 MJ/kg. They found that the water-holding capacity of the hydrochar decreased with an increase in HTC temperature, with more water being easily removed by mechanical dewatering at higher temperatures. They also assessed the potential processing of the process liquid in AD. They found that the process liquid produced at 230 °C had the highest level of chemical oxygen demand (COD) and total organic carbon (TOC) and a low ammoniacal nitrogen concentration; high levels of ammoniacal nitrogen can be problematic in AD reactors. They concluded that this was due to the majority of the decomposed organic matter in the feedstock being retained in the solid at this temperature.

Alternative methods for improving the fuel characteristics of hydrochar exist, such as the co-HTC of bituminous coal waste and food wastes studied by Mazumder et al. [78]. Up to 40% of extracted coal ends up as coal waste, with an energy content of 40–70% of that of coal. They found that the co-HTC of top bituminous coal waste with food waste resulted in a hydrochar with superior fuel characteristics. Like with Wang et al., 230 °C was found to be the optimum HTC temperature, resulting in a hydrochar with an energy content of 23 MJ/kg, and an ash content of 28 wt.%, lower than 66 wt.% found in raw coal waste. Importantly, there was a reduction in sulphur content, down to 1.4 wt.% from 8.5 wt.%. This, along with the chloride leaching, signified that the hydrochar samples would have fewer issues with corrosion and fouling. The HTC of coal waste without the presence of food waste led to little change in the properties. But, along with an increase in fixed carbon in the co-HTC hydrochar samples, there was also an increase in volatile matter, indicating that there may be issues with combustion stability.

From the previous studies detailed above, it is clear that following HTC, the concentration of sodium and magnesium in the hydrochar is lower than the original feedstock, therefore reducing the char’s propensity to slagging and fouling, While the fuel properties of the char compared to the feedstock are generally improved, the economic feasibility of combusting such a high ash material should be questioned. Even if the boiler were unaffected by the high

ash content, the removal of ash would be a costly process. This is especially true as the HHV of hydrochar tends to be lower than coal and other commonly used solid fuels, meaning higher quantities of char would have to be combusted to generate the same amount of energy.

2.4.2 Hydrochar soil application

Due to its relatively high carbon content and nutrient content, the use of hydrochar as a soil amendment is an option. The potential for hydrochar for carbon sequestration was highlighted back in 2011 by Sevilla et al. [79], who argued that its high carbon content and recalcitrant nature made it a promising candidate. They went on to propose that the chemical properties of hydrochar and water holding capacity meant that it would have a beneficial effect upon soil productivity. However, they did not conduct any plant or germination trials of the hydrochar. Additionally, there was no mention of the many potential contaminants contained in waste biomasses. Indeed, both feedstocks used in their study, eucalyptus sawdust and barley straw, are relatively clean waste streams and may not be representative of feedstocks used in HTC.

Early studies of soil application in the years following Sevilla's [79] work found hydrochar to be phytotoxic, resulting in total germination inhibition when no pre-treatment was used [80,81]. Hydrochar has a reduced stability compared to biochar from pyrolysis and so exhibits substantially different characteristics [82]. There are a number of obstacles that are preventing the acceptance of hydrochar as an adsorbent or soil improver. It is potentially acidic, has a low specific surface area, and contains polycyclic aromatic hydrocarbons (PAHs) and toxic organic substances [3]. Additionally, the true benefit of high water retention was challenged with Fornes et al. [83] discovering that hydrochar-containing substrates used in their plant trials retained too much water, causing high levels of CO_2 and issues with excess respiration in the soil. However, various biological [81,84,85], chemical [86–88], physical [80,89] and thermal [86,87] pre-treatments for hydrochar have been shown to lower the phytotoxicity of hydrochar. Both Bargmann et al. [80] and Dalias et al. [89] found that washing hydrochar reduced germination inhibition, concluding that the phytotoxic substances contained in hydrochar are water soluble. But of course, this then raises the issue of what is to be done with the contaminated rinse water generated from washing hydrochar. Busch et al. [81] explored diluting hydrochar with compost on a 3:1 ratio and found the phytotoxic effects of hydrochar to be completely eliminated, and in some cases growth was even stimulated by this hydrochar-compost mix. Other studies have shown even higher concentrations of hydrochar to have beneficial effects on plant growth. Roehrdanz et al. [84] studied co-composted hydrochars as a growing media in horticultural crops. They found that co-composting led to a higher germination rate and plant growth. Increasing the concentration of hydrochar in the compost mixture up to 50 vol.% improved the plant growth results, becoming comparable with

peat-based gardening substrate. Álvarez et al. [85] looked at using hydrochar from waste digestate alongside peat as a growth medium. Their results showed hydrochar-peat mixtures were suitable for the preparation of growing media. A 50% volume ratio mixture of hydrochar with peat increased ryegrass production by more than 180% and improved the water holding capacity. While the results of these studies seem promising, they were primarily focussed on use as growth media and so any long-term effects associated with large hydrochar concentrations were not considered.

Thermally pre-treating the hydrochar before soil application has also been investigated. Hitzl et al. [86] found that torrefaction of the hydrochar between 250 °C and 600 °C eliminated the phytotoxicity of the hydrochar, with 275 °C being the optimum temperature in terms of a trade-off between phytotoxicity reduction and solid mass loss. It was also suggested that the liquid by-product from this pre-treatment process be used as an energy source to heat the HTC process itself. Bahcivanji et al. [87] pyrolyzed hydrochar at both 350 °C and 550 °C, finding that it resulted in a higher porosity and carbon content. The germination index of the pyrolyzed hydrochar indicated that it would result in phytostimulation, with better results at the higher pyrolysis temperature. It performed favourably compared to biochar from pyrolysis alone, which resulted in phytotoxicity. An alternative method of hydrochar treatment is the addition of minerals, as explored by Mumme at al. [88]. They added zeolite, wood ash and lignite fly ash as mineral additives to raw hydrochar samples, creating biochar-mineral composites. They found that these reduced the phytotoxicity of raw hydrochar, but the biochars from pyrolysis alone still performed better. The differences in biochar findings for Bahcivanji et al. and Mumme et al. highlight the complex nature of biochar and its suitability in soils. Bahcivanji et al. used green waste from parks and gardens as the feedstock for HTC and pyrolysis, a much cleaner feedstock than the sewage sludge used in Mumme et al.'s study.

Melo et al. [90] found that the HTC of sewage sludge promoted pore structure and reduced the concentration of trace elements. However, its application to soil ultimately lead to a reduction in plant growth. Due to the restrictions of using sewage sludge on agricultural land, they compared the PAH concentration of the hydrochars to international standards. The results showed that the sum of PAH concentration fell below the maximum threshold set by the International Biochar Initiative [91], but exceeded the limit defined by the European Biochar Certificate [92]. They also conducted a survey to assess farmers' perception and acceptance of application of hydrochar from sewage sludge to their land. The results showed that younger farmers who tended to have higher levels of education were more likely to accept its use. Farmers against the use of sewage sludge-based hydrochar cited the comparison with conventional fertiliser prices and logistical costs as their greatest concern. It is important to note that the survey was carried out among Brazilian farmers, and perceptions are likely to

change with location due to the large difference in legislation and common practice across the world. In the UK, application of sewage sludge to land is permitted and regulated by a code of practice, promoting its benefits but also warning against the hazards [93]. Miliotti et al. [94], too, considered the adherence of hydrochar with biochar standards. They compared hydrochar and biochar from digestate, finding that most char properties did not exceed the limits set out in international standards [91,92].

Lanza et al. [95] considered the long-term effects of using hydrochar in soil by conducting a two-year field experiment. Their experiment concluded that raw hydrochar samples would degrade over a period of tens of years, and so could contribute to long-term carbon sequestration. Fermented samples of hydrochar contained fewer unstable compounds but overall they were less stable in the field than raw hydrochar. While hydrochar for carbon sequestration seemed promising, they highlighted the need for further investigation into the long-term effects on nutrient cycling and plant growth. Gronwald et al. [96] investigated the stability of both biochar from pyrolysis and hydrochar in soil and found that although hydrochar was unsuitable as a long-term solution, its low stability, higher reactivity and higher proportion of functional groups could prove beneficial in markets other than carbon sequestration, such as a slow-release fertiliser. Idowu et al. [97] is one of the few studies so far to investigate the market potential for hydrochar as a fertiliser replacement, using food waste as an example HTC feedstock. When studying the nutrient availability through leaching experiments, they found that in the short-term nitrogen release was small but almost all the phosphorous was released from hydrochars produced at <250 °C. This led to predictions that for the US market, approximately 1.0% and 2.3% of nitrogen and phosphorous-based fertilisers respectively, could be replaced by nutrients in the hydrochar and process liquid if all food waste produced in the USA was diverted from landfill to HTC.

2.4.3 Process Liquid

Currently, standard industrial practice is that the process liquid product from HTC is either recycled back and mixed with the original feedstock to improve its pumpability, or spread to land for crop irrigation [98]. Recirculation of process liquid has been a key area of HTC research during the past decade, particularly for HTC feedstocks with lower moisture contents. In industrial processing, when the moisture content is too low for easy pumping of the feedstock, water is added and mixed with the feedstock. Therefore, recirculation offers a reduction in water use and waste disposal. On top of this, research suggests that process water recirculation can lead to improvements in the HTC process and product quality. In 2013, Stemann et al. [68] concluded that process water recirculation concentrated and catalysed dehydration reactions during HTC. This led to an increase in the carbon and energy contents

of the hydrochar as well as enhancing its dewaterability. They observed a polymerisation reaction too, with the formation of an additional solid substance from compounds in the process liquid. Uddin et al. [99] came to similar conclusions that same year, measuring an increased hydrochar yield of 5–10 wt.%. However, they noted the change in hydrochar energy content and hydrophobicity as negligible. Other studies have observed a similar increase in hydrochar yield, carbon and energy contents, as well as a decrease in its ignition temperature and reactivity [100–103]. In addition to the improvement in hydrochar yield and quality, Wang et al. [104] found that recirculation upgraded the process liquid too. Seed germination tests suggested a partial alleviation in phytotoxicity, and biomethane potential (BMP) tests indicated a boost in methane production for diluted process liquid samples.

Harnessing the carbon content of the process liquid through anaerobic digestion (AD) for biomethane production has gained increased interest in recent years. Wirth and Mumme [105] ran HTC process liquid in an anaerobic digester for a hydraulic retention time of 42 days. The results showed process liquid to be suitable for use in AD, supported by the results by a number of other studies [106,107]. Researchers have gone on to assess the effect of HTC severity, finding that lower HTC temperatures tended to improve the biomethane potential (BMP) of process liquid [108–110]. Some studies have gone on to consider the AD of the hydrochar alongside the process liquid. Zhao et al. [111] used food waste in HTC and concluded that adding the hydrochar to the process liquid before AD could increase the specific methane yield by a factor of 2.5 compared to when no hydrochar was added. Brown et al. [112] used macroalgae as feedstock for HTC. They found that the combination of HTC and AD of the process liquid resulted in a higher energy output, of up to 172%, compared to the AD of untreated macroalgae. However, they still concluded that hydrochar would produce more energy as a solid fuel rather than in AD.

An alternative to recirculation or AD of process liquid is use on land or discharge to the watercourse. An analysis by Becker et al. [113] highlighted the potentially toxic nature of untreated process liquid, having qualities harmful to aquatic life. The findings of Fregolente et al. [114] supported this as well as reporting that concentrated process liquid samples inhibited the germination of various crops. However, diluted samples appeared to stimulate growth of both maize and tomato. Reza et al. [115] investigated wet air oxidation as a treatment for process liquid, a process in which excess air/oxygen dissolves into the process liquid at high temperature and pressure and reacts with the organic substances to produce CO_2 and clean aqueous stream. They reported the resulting aqueous stream was clear and consisted of non-toxic compounds, and so the wet air oxidation treatment was judged successful. Microalgae cultivation in the process liquid as a treatment method was explored by Belete et al. [116]. They found that nitrogen, phosphorous and carbon contents all reduced by >80% after 6 days,

resulting in process liquid of suitable quality for reuse or discharge. Phosphorous is a high-value element, with interest in its recovery for economic purposes rather than solely as a way to reduce the harmful nature of the process liquid. Crossley et al. [117] measured a 75% extraction of phosphorous from the process liquid using microfiltration. Ultimately, the level of treatment required for the process liquid will depend on local restrictions and methods of disposal. While recirculation, AD, treatment and nutrient recovery of the process liquid can all offer improvements in energy generation and high-value products, the economic attractiveness of these will heavily depend on the HTC feedstock itself and local market conditions.

2.5 Techno-economics

While understanding the HTC process is important for its development, a consideration of the techno-economics gives a better understanding of the industrial viability of the process. For example, in industrial HTC settings, the operating conditions are likely to be governed by external factors rather than the theoretical optimum as found by reaction models. An increase in temperature generally leads to a higher degree of carbonisation and thus a higher value hydrochar. However, if the increase in hydrochar sale price does not offset the increase in operating costs, the operator may choose a lower process temperature. Similarly, operating at longer residence times can improve the quality of hydrochar, but would invariably lead to larger plant equipment. There would be a trade-off between improved products and higher capital costs. Additionally, the operator may choose not to alter the initial moisture content of the feedstock, despite potential improvements to the process or products, in order to save on the cost of dewatering or water addition.

HTC is still a developing technology, but there are a number of pioneering companies operating at industrial scale. These include: Antaco (UK), C-Green (Finland), HTCycle (Germany), Ingelia (Spain), GRENOL (Germany), TerraNova Energy (Germany) and SunCoal Industries (Germany). These companies generally have specific agreements to treat waste from local authorities or organisations. The treatments of sewage sludge, paper mill sludge or green waste are the most common. Therefore, there is a need to explore the techno-economics of a greater range of HTC processes to better understand its viability. A model developed to be suitable for easy adaption to feedstock variables and location specific aspects would aid this.

2.5.1 Energy

Due to the elimination of a feedstock drying stage, HTC has often been championed as a lower cost, more energy efficient thermochemical treatment for wet biomass compared to

more established techniques such as pyrolysis. Table 2.2 shows a series of studies that have looked at the energy efficiency of HTC and whether, through the combustion of the resultant hydrochar product, the system is self-sufficient. The positive net energy values indicate systems which have the potential, or claimed potential, to be energy self-sufficient.

Table 2.2: Studies of the net energy production from HTC.

Feedstock	Experimental moisture content (wt.%)	HTC temperature (°C)	Hydrochar yield (% d.b.)	Hydrochar HHV (MJ/kg d.b.)	Net energy (MJ/kg dry feedstock)	Ref.
Poultry litter	75	250	46	24	8	Mau and Gross [118]
Recycled paper sludge	10–50	180–250	95–60	22–26	5–8	Mäkelä and Yoshikawa [119]
Food waste	75	200	61	30	12 (incl. AD of process water)	Gupta et al. [120]
Yard waste	91	160–200	79–46	16–25	15–5	Sharma et al. [121]
Sewage sludge	95	200	67	18	4 (15% solids) – 15 (25% solids)	Danso-Boateng et al. [122]

Energy efficiency studies have shone light on best practices for HTC operation. Danso-Boateng et al. [122] proved the energetic benefits of using feedstock with a lower moisture content. Zhao et al. [123] suggested that implementing mechanical dewatering of the products improves the thermal efficiency of the process. Afolabi et al. [124] studied the HTC of spent coffee grounds. They predicted that with a HTC process operating at 216 °C and 1 hour, hydrochar with a yield of 64 wt.% and an energy content of 31.6 MJ/kg could be produced. Therefore, if all 500,000 tonnes of spent coffee grounds generated annually in the UK were processed with HTC, the combustion of the resulting hydrochar could replace over 5% of the bioenergy used for electricity generation in 2019 [125]. Their study did not consider the energy consumption of the HTC plant itself and was only based on the energy content of the hydrochar. Without detailed modelling or validation, the robustness of this result is brought into question.

These studies highlight the relative energy efficiency of HTC as a treatment for wet waste and the potential of hydrochar for energy generation. The basic principles of the energy balance models in these works have been developed further by those exploring the economics of industrial-scale HTC plants.

2.5.2 Economic

There remains the need for a validated economic model of an industrial-scale HTC plant. Stemann et al. [126] investigated the industrial-scale HTC of palm oil fruit bunches in South East Asia for the production of hydrochar. They modelled two plants, one with a capacity of 5.7 tonne/hour and a larger one processing 13.8 tonne/hour. They calculated the total capital investment to be 9 million € and 16.5 million € respectively. Including the cost of hydrochar shipment to Europe, the hydrochar production cost came to between 7.9 €/GJ and 9.7 €/GJ. With an HHV of 27.4 MJ/kg, this equates to a hydrochar cost of 217–266 €/tonne (£194–237/tonne). Compared to coal prices, at roughly £50/tonne, this was deemed too high, and it was argued that public policy incentives would be required for an economically efficient production of hydrochar. This conclusion was echoed by Meisel et al. [127]. They considered the economic potential of the HTC of sewage sludge in Germany and found that the additional cost of HTC was not offset by the increase in agricultural yield, if the hydrochar were to be applied to land, or the increase in energy production, if it was used as a fuel. Therefore, HTC was not found to be favourable compared to current standard practice. However, the legislation in Germany is set to change in 2029 and 2032, ruling out the direct application of sewage sludge to land. Under these circumstances, HTC could serve as a useful mitigation tool for paying higher disposal costs. The greenhouse gas (GHG) savings associated with HTC were also measured. However, it was found that HTC did not offer substantial improvements. The most promising of the techniques studied was the dewatering and anaerobic digestion of sewage sludge. Under the new legislation due to be brought in, the question of its disposal would still remain however.

Other studies have given much more optimistic production costs for hydrochar. Shao et al. [128] performed a basic cost analysis for the microwave HTC of green waste and calculated a hydrochar price of $50/tonne (£38/tonne), lower than the price of coal. However, this analysis did not cost an industrial-scale microwave HTC plant and used assumptions from laboratory-scale experiments. Nakason et al. [129] also performed basic cost calculations, finding hydrochar as a fuel production to be profitable, but without detailing the size of plant, plant equipment or even the thermal energy use. Kempegowda et al. [130] considered integrated hydrochar and activated carbon production in rural communities. They concluded that the process was technically and economically feasible, but the model was based on the simplistic assumption that hydrochar-water slurries would combust in diesel engines as biochar-water slurries have been shown to do in a few limited studies. However, hydrochar acts differently in water to biochar and no consideration of the availability, type or cost of the raw feedstock was made. Unrean et al. [131] compared the economic viability of HTC, pyrolysis and AD of

rice husk. They reported HTC to be the most economically and environmentally promising, largely due to its high solid-operating capacity. While true that HTC can process wet wastes with higher moisture contents, the solid-operating capacity would be the same for a pyrolysis system treating the same quantity of raw feedstock. It seems as though there was an error in mass loading calculations and so a true comparison between the processes was not in fact made. Tradler et al. [132] considered a 50 litres, decentralised HTC unit treating restaurant food waste. Their costs were calculated based on small-scale boiler technologies and they found that the system would become profitable in its eighth year of operation. While being an interesting concept, no considerations were made for how well or what restrictions there would be on the combustion of hydrochar in a decentralised system. There is clearly scope to look at this idea further.

Of the most comprehensive economic HTC assessments, results are mixed. Saba et al. [133] modelled a system based on the co-HTC of a coal-miscanthus blend at an industrial-scale. Sizing and costing of plant equipment was performed, and the breakeven selling price of the hydrochar was calculated as $117/ton (£89/tonne), coming in as low as $106/ton (£81/tonne) in the sensitivity analysis. The largest contributor to the operating costs was the feedstock itself, accounting for 73% of the manufacturing cost. They calculated that the cost of producing electricity in this co-HTC plant was nearly twice the cost of electricity from standard coal-fired plants. Mazumder et al. [134] performed a similar study, this time looking at the co-HTC of coal waste and food waste. Using lower-value feedstocks lowered their hydrochar production cost to $60–69/tonne (equivalent to £46–53/tonne). Their sensitivity analysis revealed feedstock purchasing cost and transportation costs to be the most influential variables. Both of these studies support the case for using very low-value, wet feedstocks in HTC. They also brought into question whether using coal as a feedstock for a co-HTC process, with the aim of producing a high value hydrochar suitable as a fuel, was economically feasible. Lucian et al. [135] modelled the HTC of off-specification compost and grape marc on an industrial scale. They considered different HTC temperatures and times in their analysis and concluded that processing grape marc at 200 °C for 1 hour gave the best economic results. They calculated the hydrochar production at 157 €/tonne (£140/tonne), with a break-even point of 200 €/tonne (£178/tonne). These costs are in the region of predicted hydrochar prices in Europe and therefore they concluded that HTC is an attractive process for producing a CO_2 neutral biofuel [136]. The model created by Lucian [135] was thorough, detailing the product flows and each part of the process. However, it was still based on experimental results and literature findings, lacking validation from industry. The labour costs, in particular, do not seem to account for the true number of shift patterns and operators required at a continuously operating plant.

The assumptions, modelling and results of these techno-economic models are all fairly inconsistent. Generally, the most promising results are from studies where modelling was performed in very little detail. This is presumably due to the glaring omission of critical HTC plant equipment. The production cost of hydrochar tended to increase as the complexity of the model and depth of analysis increased. Models using relatively high-value feedstock resulted in high hydrochar production costs [133,134]. The economic viability of models using lower-value feedstocks depended on the assumptions made, with some finding hydrochar production costs matching current market prices [135], and others concluding HTC is not currently financially feasible [126,127]. Current techno-economic models lack flexibility and proper validation. This field of research would benefit from an exhaustive HTC process and economic model, focussing on low-value wastes and backed up by industrial plans and data. This would lead to more informed decisions on the technical and economic feasibility of HTC at scale in a range of markets.

2.6 Conclusions

HTC research has increased rapidly over recent years, with investigations focussing on the reaction itself as well as its products. An increase in HTC process severity, specifically the temperature and time, leads to an increase in degree of carbonisation. This correlates to a lower yield of hydrochar, yet an increased carbon and energy content (HHV). This has been successfully modelled for specific feedstocks by various researchers. Universal models for all feedstocks have been generated but they have a limited capability in the accurate prediction of HTC product yields and properties. It has been reasoned that they would be better used as a screening tool before developing a feedstock-specific model for greater depth.

Hydrochar has proven to be a suitable fuel product in certain circumstances. Generally, these have been HTC at high temperatures of lignocellulosic feedstocks with low ash contents. The minimisation of ash is essential in producing a solid fuel in order to reduce its propensity for slagging and fouling in a boiler. Soil application has been proposed as an alternative use of hydrochar, whether with the aim of carbon sequestration or as a fertiliser. The majority of studies agree that untreated hydrochar has issues with phytotoxicity and stabilisation, however numerous biological and thermal treatments have been shown to improve these properties. Although a small number of large-scale, long-term field trials have been completed, more are necessary to better understand the stability of hydrochar in soil. The process liquid is also a valuable HTC product and the effect of recirculation or its use as a substrate for AD has been discussed. Generally, process liquid recirculation leads to higher yields of hydrochar with an increased carbon content. The AD of process liquid has given promising results, indicating a potential to increase the overall energy-efficiency of HTC.

Various studies have calculated HTC to be an energy-efficient process, with the energy contained in the products outweighing the energy required to run the reactor. There are a number of techno-economic models predicting the capital and operating cost of HTC and calculating the break-even selling price of hydrochar. The results of the most developed techno-economic models are inconsistent, highlighting the need for industrially validated case-studies of HTC at a large scale. Additionally, the cost of the feedstock has been shown to have the largest effect on the profitability through sensitivity analyses. This suggests that HTC processes targeted at very low-value waste products would be the most economically viable. This is supported by current industrial HTC operators tending to have contracts with local authorities or companies to treat a particularly problematic waste in that area. If the number of HTC plants were to increase, it is expected that these contracts and the feedstocks used would diversify, and so a robust estimate of process costs as well as feedstock costs would prove useful.

Overall, HTC shows potential for the treatment for wet waste, both technically and economically. However, much more needs to be understood of real-world applications and economic predictions in order to assess its viability at large-scale.

3 Methodology

3.1 Introduction

This chapter details the methodology used during the experimental, modelling and market analysis work. Data was collected experimentally, from literature and industrial designs and was used to create a process model of an industrial-scale HTC facility. The results of the process model were then used to analyse the potential of HTC of digestate in the UK. The process followed is summarised in Figure 3.1.

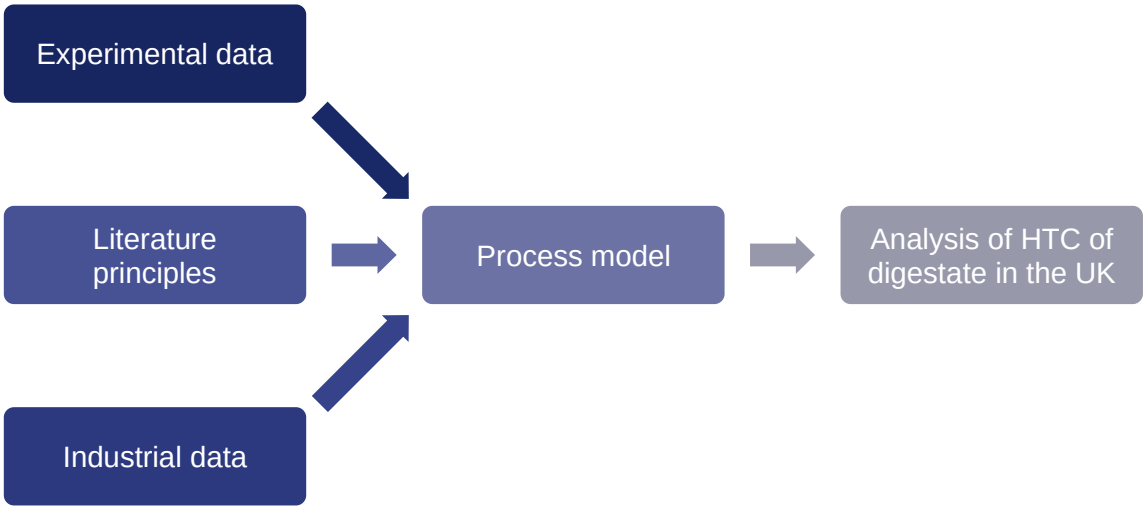


Figure 3.1: A flow diagram of the process followed.

The methods followed in each of the steps were as follows:

- Three different types of feedstock were used experimentally in HTC to represent the most common forms of digestate produced in UK AD plants.
- They were fed into a laboratory-scale HTC reactor at a range of temperatures, residence times and moisture contents.
- The yields and HTC product compositions were measured and regression models were generated.
- A flexible process model was then created of an industrial-scale HTC plant based on the design of an existing plant.
- The regression models were fed into the process model and were used to predict product yields and compositions depending on the HTC conditions.
- Cost models from literature were added to the model which were then validated using the industrial data.
- A market analysis was then performed based on the HTC product properties, cost estimates and market predictions.

3.2 Experimental Procedure

The experimental work comprised of three main stages: laboratory-scale HTC, sample analysis of the feedstock and HTC products, and germination and plant growth trials using the hydrochar products. The results of these steps were used later in the process modelling and market analysis work.

3.2.1 Feedstock Use

In 2019, 12.5 million tonnes of feedstock were processed by 486 anaerobic digestion (AD) plants resulting in over 500,000 dry tonnes of digestate production [15,16]. The number of AD plants, and therefore amount of digestate produced, is rising [19]. Experimental work focussed on 3 sources of digestate, namely food waste, farm slurries and maize crop from different AD operators. These were selected to represent the variety of digestate produced in the UK and to explore the range of markets for the resulting hydrochars. In May 2018 there were 83 AD plants processing food waste, 167 processing farm slurries and 139 processing maize crops [15]. In total, these plants processed over 8 million tonnes of feedstock a year, producing in excess of 7 million tonnes of whole digestate. Details of the digestate sources are shown in Table 3.1.

*Table 3.1: Types of digestate used experimentally. *Digestate production is estimated assuming a digestate yield of 87%, given as the average conversion by WRAP* [16].

	Waste-based digestate	**Farm-based digestate**	**Crop-based digestate**
Developer	Langage Farm, Devon	JF Temple & Sons, Copys Green Farm, Norfolk	Severn Trent Water, Bulcote Farm, Stoke Bardolph, Nottinghamshire
AD feedstock	Food waste (hospitality, household, retail and factory)	Dairy cow slurry, maize silage and whey	Maize silage
Biomass input to AD plant (tonne/year)	20,000	5,000	79,000
Whole digestate production (≈5 wt.% solids) (tonne/year)*	17,400	4,350	68,730

3.2.2 HTC Procedure

Three factors were varied during the HTC runs: temperature (180–250 °C), residence time (60–180 minutes) and moisture content (80–95 wt.%) [137]. The upper and lower limits of these process conditions were set to represent the mid-range of values typically found in other

experimental HTC studies and industrial scale operation [65,98]. Each HTC run was carried out in a 75 ml Parr reactor heated by a fluidised sand bath. 3 g of dried feedstock was added to the reactor and the corresponding volume of distilled water was added to achieve the desired moisture content. The reactor vessel was flushed with 1 bar of nitrogen multiple times to remove air from the reactor and to ensure there were no leaks. The nitrogen in the reactor was vented and its volume measured each time. Once measurements were consistent, 1 bar of nitrogen was added for a final time and the reactor was sealed. Once the sand bath had been heated to reaction temperature, the Parr reactor was lowered inside the sand bath and held there for the desired residence time. As the reactor was not pre-heated before entering the sand bath, a temperature drop of roughly 20 °C was measured immediately after entry. The temperature control was adjusted to minimise the heating time of the reactor, while ensuring that the desired reactor temperature was not exceeded. This process was followed for each of the reactions carried out. The experimental setup is pictured in Figure 3.2.

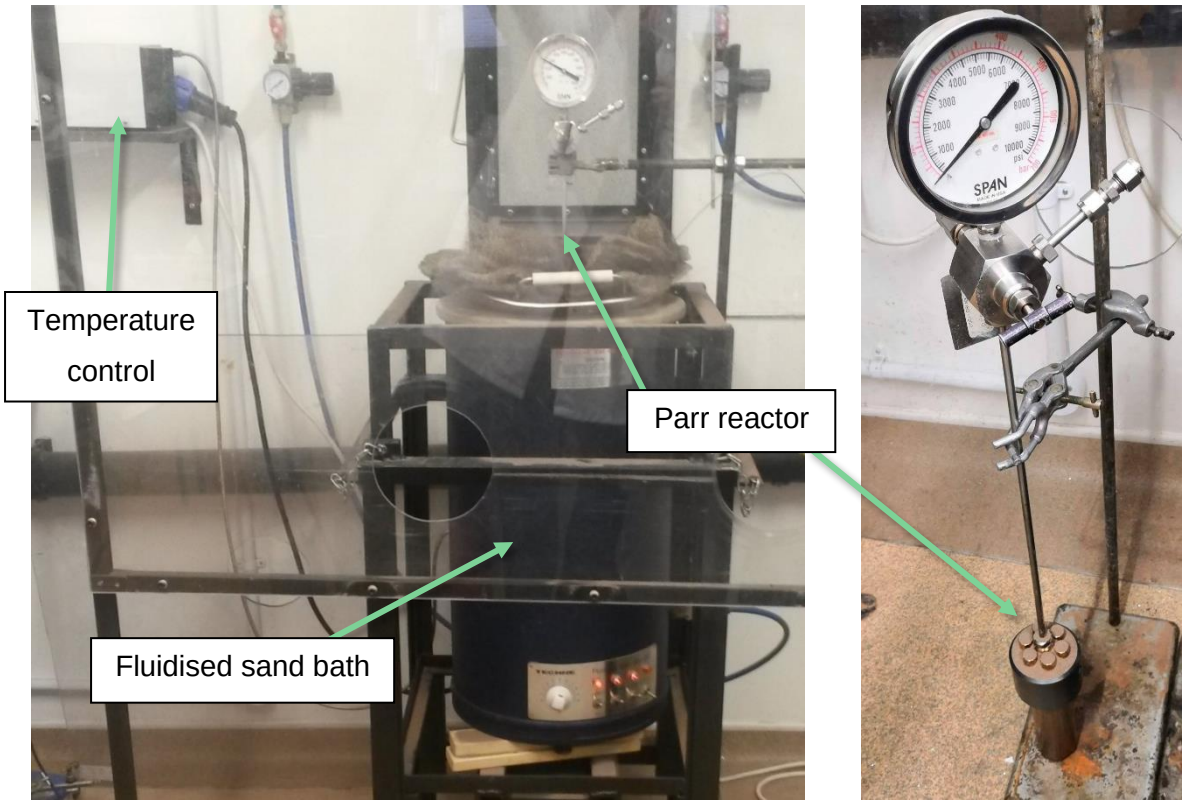


Figure 3.2: Reactor and sand bath assembly.

Once the residence time had passed, the reactor was removed from the sand bath and allowed to cool down to room temperature. The volume of the process gas (PG) was measured using a glass syringe and recorded. The composition of the process gas was assumed as 100% CO_2, based on information from industrial HTC operators, and its mass was calculated using the ideal gas law and its molecular weight (Eq. 3.1).

$$Mass\ of\ PG\ (g) = \frac{PG\ recovered\ (ml) - N\ added\ (ml)}{molar\ volume\ of\ a\ gas\ (cm^2)} \times Molar\ mass\ CO_2\ (g/mol) \qquad \text{Eq. 3.1}$$

A sample of the process liquid (PL) was separated using a vacuum filter and set aside for analysis. The rest of the reaction products, consisting of hydrochar (HC) and the remaining process liquid and water, were washed using distilled water and placed in an oven at 105 °C. The washed hydrochar was dried overnight and weighed to find the dry hydrochar mass yield. Using the measured mass of dry hydrochar and calculated mass of process gas, the mass yield of process liquid was determined by mass balance (Eq. 3.2).

$$Mass\ of\ PL = Mass\ of\ wet\ feedstock - Mass\ of\ PG - Mass\ of\ dry\ HC \qquad \text{Eq. 3.2}$$

3.2.3 Sample Analysis

Thermogravimetric analysis (TGA) was used to measure the moisture content, volatile matter, fixed carbon and ash content of the digestate and hydrochar. The International Organisation for Standardisation method for coal (ISO 17246:2010) was followed where the moisture content was determined at 105 °C; the volatile matter content at 900 °C; the ash content after burning at 500 °C and then 815 °C; and the fixed carbon content was calculated by difference. The TGA measurements were performed in triplicate using a Discovery TGA manufactured by TA Instruments. Figure 3.3 shows the heating profile used.

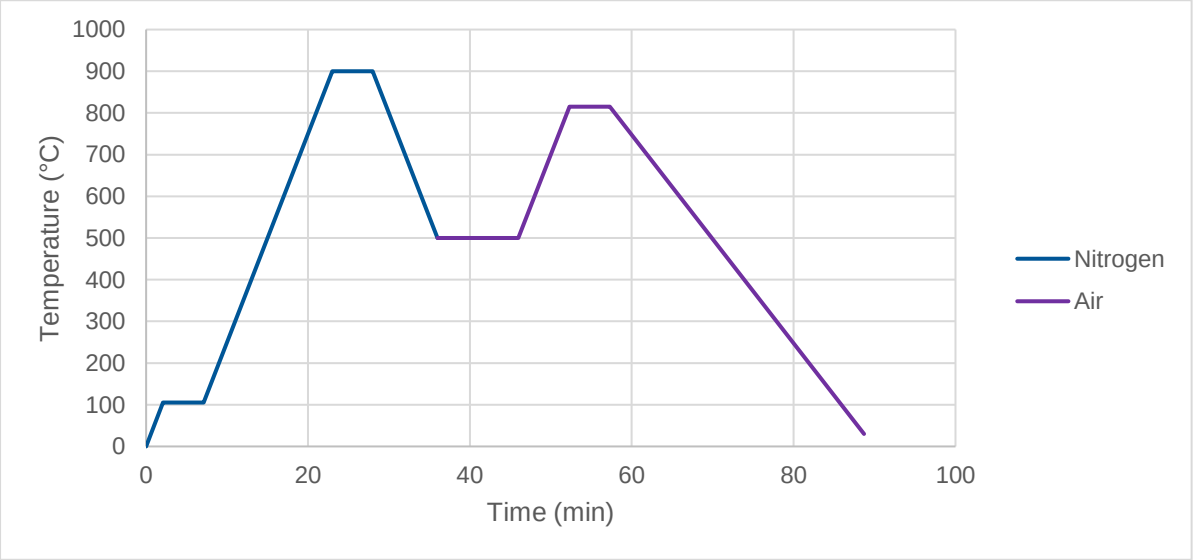


Figure 3.3: Heating profile used for proximate analysis.

Ultimate analysis was performed using the CHN628 Instrument from LECO Corporation. The weight percentages of carbon, hydrogen and nitrogen of the feedstock, process liquid and hydrochar samples were determined using the CHN628. The oxygen content was calculated by difference. Sulphur content was not measured and was assumed to be negligible.

The elemental mineral concentrations of the feedstock, process liquid and hydrochar samples were determined in accordance with BS EN ISO 16967: Solid biofuels — Determination of major elements. The process liquid was filtered through a 5 µm syringe filter to remove any solid particles that would interfere with the analysis. It was then diluted by a 1 to 10 acid dilution for analysis. For analysis of the elements potassium, manganese and sodium, a further 1 in 10 dilution was required to bring the values to within the calibration range. A CEM MARSX sealed vessel microwave digestion system was used for the total digestion of the hydrochar and feedstock samples. An Agilent Technologies 5110 VDV ICP-OES instrument was used for determination of elemental concentrations of the digested biofuels.

The chemical oxygen demand (COD) of the process liquid samples was determined using HACH Lange cuvette test kits (LCK014) in combination with a visible spectrophotometer that averaged 10-fold absorbance readings. The process liquids were prepared with a 1 in 10 dilution.

3.2.4 Germination and plant growth trials

Hydrochar produced from food-waste digestate was used to investigate the suitability of hydrochar as a soil application for either carbon sequestration or fertiliser use. Plant trials were performed to investigate the phytotoxic properties of the hydrochar and whether its addition to plant growth media would inhibit germination, thus ruling out its use in soil application. The trials were contracted out to the Biorenewables Development Centre (BDC), an open-access R&D centre based at the University of York. Hydrochar samples were sent to BDC and they performed the plant growth trials using their growth cabinets and analytical equipment. Due to the large quantities of hydrochar required for plant trials compared to in the laboratory HTC test runs, hydrochar was sourced from the demonstration-scale HTC plant at CPL Industries. CPL Industries had been recently been using food-waste digestate to produce hydrochar. The food-waste digestate was provided by GWE Biogas, an AD facility processing 50,000 tonnes of food waste annually and producing somewhere in the region of 43,500 tonnes of whole digestate or 8,700 tonnes of dewatered digestate fibre (25 wt.% solids). The digestate was dewatered before entry to the HTC plant which operated under standard industrial conditions of 200–220 °C for a residence time of several hours, including heat-up time [98].

Prior to the germination trials, the hydrochar was subjected to further thermal processing. This has been shown to be an important step in removing phytotoxic compounds contained in raw hydrochar [86,87]. The hydrochar was pyrolysed at 275 °C for 4 hours using a heating rate of 3 °C/minute. The treatment was conducted under vacuum to prevent any oxygen from entering the system.

For the germination trials, coconut fibre growth medium (coir) was used alongside hydrochar in different concentrations. The set of conditions used represent those can be found in other studies considering the potential of hydrochar in horticulture [83,84]. Table 3.2 shows the ratios used.

Table 3.2: Volumes of coir and biochar required for plant growth trials.

Ratio		**Amount Needed (l)**	
Coir	Hydrochar	Coir	Hydrochar
100	0	3.0	0.0
90	10	2.7	0.3
80	20	2.4	0.6
70	30	2.1	0.9
50	50	1.5	1.5
0	100	0.0	3.0

Each mixture was distributed equally between three plant growth tray inserts. The bottom of each insert was covered with a clean plastic bag to prevent water run-off being taken up by neighbouring inserts when resting in plant growth trays.

Mixtures were equilibrated overnight in a Panasonic MLR-352 PE plant growth cabinet set to 22 °C, before basil seeds were sown, evenly spaced at a rate of 25 seeds per insert. All inserts were watered with 200 ml and the inserts were set in trays and distributed randomly across the shelves of the growth cabinet. The growth cabinet was set to 22 °C for 16 hours a day.

Germination numbers were scored at the end of one and two weeks after sowing. The number of seeds germinated and any observations relating to the health or growth of the plants or growth medium were recorded. Seedlings were then thinned to 10 per insert and incubated in the growth chamber for a further 6 weeks under the same conditions. Further readings were taken during and at the end of the 8-week period. At the end of the experiments, the basil plants were cut and dried at 105 °C to determine the dry weight.

3.3 Statistical Analysis of Results

Regression models for the product yields and properties were developed for each of the feedstocks investigated. The range of process conditions were selected to match those typically found in industry and other statistically based models investigating hydrochar properties [98,137].

For the waste-based digestate, a Box-Behnken design of experiments was carried out to investigate the effect of three variables: time, temperature and moisture content. The Box-Behnken model uses treatment combinations at the midpoints of the edges of the experimental space, a visualisation is shown in Figure 3.4. It has fewer design points and therefore is often

less expensive to run than central composite designs. Additionally, all design points are inside the set minimum and maximum limits of each factor. This ensures that all runs remain within the range of interest and safe operating zone.

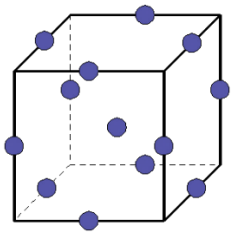


Figure 3.4: Box-Behnken design of experiments using 3 factors.

The Box-Behnken design has been used in literature studies on the effect of process conditions on the HTC of digestate, giving results with a good fit [40,43]. Following the completion of these experiments, the centre-point run was repeated a further 2 times, resulting in 5 experimental runs under the same conditions to assess repeatability. The order of the 15 runs was randomised to reduce the effect of bias and is shown in Table 3.3. The Response Surface Design on Minitab was used to analyse the results. The backwards elimination method was used to remove any variables that had a significance of less than 0.05.

Table 3.3: Process conditions and run order of HTC experiments.

Run no.	Temperature (°C)	Time (minutes)	Moisture content (wt. %)
1	215	120	87.5%
2	180	120	80.0%
3	215	120	87.5%
4	215	60	95.0%
5	215	180	95.0%
6	250	180	87.5%
7	250	120	95.0%
8	215	120	87.5%
9	215	180	80.0%
10	215	60	80.0%
11	250	60	87.5%
12	250	120	80.0%
13	180	180	87.5%
14	180	120	95.0%
15	180	60	87.5%

For the farm-based and crop-based digestates, only temperature and time were varied rather than a complete Box-Behnken design of experiments due to limitations in digestate access. A simple regression model was fitted using Minitab, based on those two variables using the backward elimination method with a minimum significance of 0.05.

3.4 Process Model

A process model for a HTC plant was created based on designs from literature and specifications of an industrial-scale facility [122,135,137]. Non-public data was provided from the demonstration plant commissioned by Ingelia for CPL Industries in conjunction with the University of Nottingham. This data included plant equipment costs, process operating conditions and overall mass balances. The plant design is modular in nature, with the number of reactors dependant on the throughput. All sizing, energy usages and costs were calculated independently using engineering principles and publicly available data. The model was then validated using confidential data from both the demonstration plant and other business models developed by Ingelia.

A flow diagram showing the different stages of the plant model is shown in Figure 3.5. The furnace was included as an optional unit operation, depending on whether upgraded hydrochar for land application was desired. The model was designed to use feedstock properties and experimental data as inputs, generating estimates for hydrochar production, energy use and capital and operating costs. The process model results were then used as the basis for the financial and feasibility analysis.

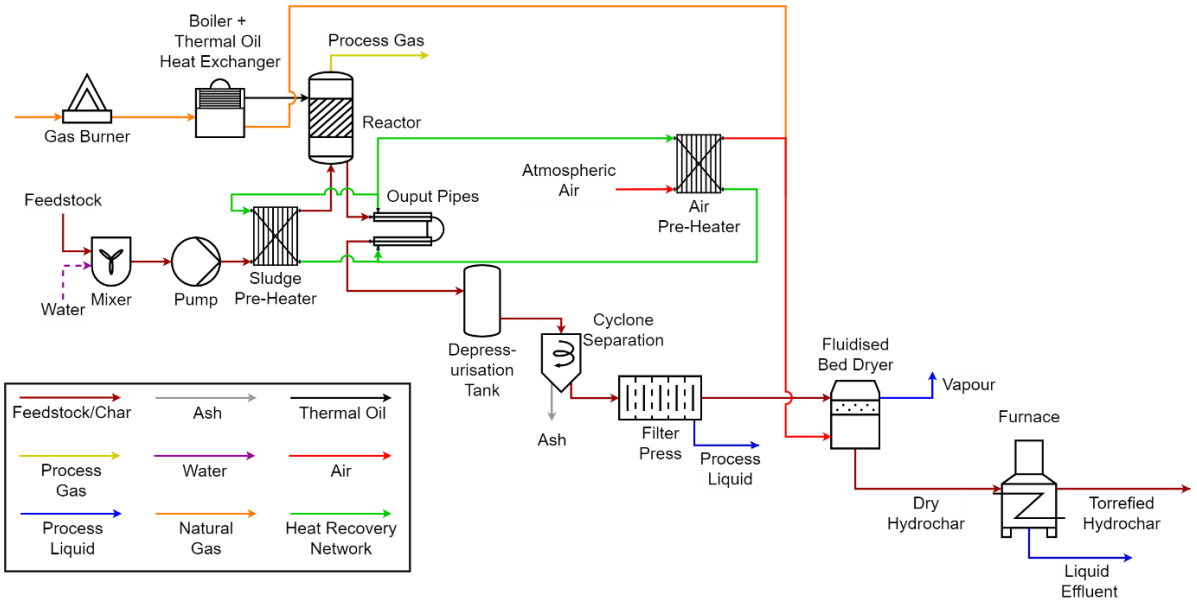


Figure 3.5: Flow diagram of the HTC plant studied. The furnace unit was optional.

Each of these unit operations was modelled as MATLAB® functions which were run together in one MATLAB® script. The model had a number of variables including type of feedstock,

annual throughput, feedstock moisture content, reaction temperature and reaction time. It was flexible and could be run either at a single design condition or over a range of conditions. When a range of conditions was investigated, a meshgrid was used so that temperature, time and moisture content were analysed simultaneously. By default, the model was written to run at 20 equally spaced values for each variable, resulting in 8000 sets of conditions.

Using the MATLAB App Designer, a custom application was created to run and display the results of the model. The app was interactive and so the user was able to define the conditions of the plant and view how this affected the various outputs. More details are given in Chapter 5 HTC Plant Model Section 5.2.1 Process model application.

3.4.1 Determination of Properties

Where measured amounts were not available, certain feedstock, slurry and hydrochar properties were estimated.

Specific heat capacity (cp) was calculated based on proximate analysis fractions and temperature [138]:

$$c_p = x_{Mois.}(4180) + x_{FC}(690 + 2.85\mathrm{T} - 0.00176\mathrm{T}^2) + x_{PVM}(1650 + 3.39\mathrm{T}) + x_{SVM}(2970 + 3.39T) + x_{Ash}(753 + 0.589\mathrm{T}) \quad \textit{Eq. 3.3}$$

where x is the mass fractions of the various components, T is the temperature in Celsius, and PVM and SVM are the primary and secondary volatile matters respectfully; any VM in excess of 10% is classed as primary volatile matter.

The higher heating value (HHV) of the digestate feedstock was calculated based on the elemental composition and ash content [139]. This was shown as the most suitable correlation for biomass feedstocks [140]:

$$HHV_{feed} = 0.3491C + 1.1783H + 0.1005S - 0.1034O - 0.0151N - 0.0211Ash \quad \textit{Eq. 3.4}$$

Density (ρ) was calculated based on the temperature and total solids of the stream [141]:

$$\rho(T, TS) = \frac{\rho_{H_2O}(T)}{1 - C(T) \times TS} \quad \textit{Eq. 3.5}$$

where ρ_{H_2O} is the density of water at temperature T, TS is the total solids content of the sample and the coefficient C is calculated by:

$$C(T) = -1.877 \times 10^{-6} \times T^2 + 1.097 \times 10^{-3} \times T - 0.156 \qquad \textit{Eq. 3.6}$$

where T is the temperature in Kelvin.

3.4.2 Feedstock Input and Dewatering

The first step of the model was to define the type of feedstock to be considered and the quantity to be processed. The amount of feedstock to be treated by HTC depended on the scale of the plant and findings from market research. A range of values were considered during the economic analysis. It was assumed that the AD process gave a digestate yield of 87% [16]. The input temperature for the feedstock mixture was set to 9.5 °C as the average annual outside temperature in the UK [142]. For digestate feedstocks with a moisture content of lower than 95%, dewatering was modelled before entry to the HTC plant. The density of the feedstock was estimated based on the total solids content and the density of water in order to calculate the volumetric flowrate entering the plant [141].

Technical and economic estimates for the dewatering of digestate were taken from the reports by Federal Ministry for Sustainability and Tourism (BMNT) in Austria and the Waste and Resources Action Programme (WRAP) in Wales [20,22]. Both studies considered a centrifuge for the dewatering of whole digestate, producing digestate fibre and digestate liquor fractions. The recovery of organic N, ammoniacal N, P and K in the digestate fibre were estimated using linear interpolation of the results from Figure 5-8 of the BMNT report. The solid recovery in the digestate fibre was estimated using liner interpolation of the results in Table A2-3 reported by WRAP. WRAP investigated the capital and operating costs of dewatering food waste whole digestate. Central Case 1 from the WRAP study was used as the digestate moisture content was closest to that used experimentally. Scenario 5 where the digestate liquor was subjected to biological oxidation using SBR and discharged to watercourse gave the best economic performance for Central Case 1. The scale of dewatering was shown to be economically insignificant. Therefore, values for Central Case 1 under Scenario 5 of an AD plant size of 25 ktonnes/year were used. All values in the WRAP report were listed as £/tonne feedstock input to the AD plant. Therefore, they were converted to £/tonne whole digestate for the centrifuge cost and £/tonne digestate liquor for the biological oxidation and discharge to watercourse cost.

3.4.3 Biomass Feeding Equipment

The digestate is fed through a biomass hopper and then a mixer to form a homogenous feedstock. The ratio of impeller diameter to vessel diameter (d/D) was set as 0.585 [143]. The ratio of the height to vessel diameter was 1.1 and mixing speed was 70 rpm as these are

common values often seen with digestate mixing [135]. Mixing time was set to 5 minutes as this was assumed to be sufficient time to create a slurry [144]. Mixer volume was calculated using the volumetric flow rate of feedstock over 5 minutes running time. Geometrical ratios were used to size the mixer.

Non-Newtonian flow behaviours were assumed for digestate mixture in which the viscosity is not constant and depends upon the shear rate of the system:

$$\mu_a = \frac{\tau}{\dot{\gamma}} \qquad \textit{Eq. 3.7}$$

where μ_a is the apparent viscosity, τ is the shear stress and $\dot{\gamma}$ is the shear rate.

For a paddle impeller, the average shear rate can be calculated assuming that the shear rate is directly proportional to impeller speed [145]:

$$\dot{\gamma} = kN \qquad \textit{Eq. 3.8}$$

where N is the rotational speed and k is a constant = 11.

A prediction of shear stress was made using Mbaye et al. where the flow properties of a number of anaerobically digested wastes were presented [146]. They found that when the solids content of the feedstock was over 10 wt.%, the digestate exhibited properties in line with the Herschel-Bulkley model whereby a yield stress must first be overcome before any flow will occur. The parameters of digested waste “A1” were taken and used here.

$$\tau = \tau_c + K\dot{\gamma}^n \qquad \textit{Eq. 3.9}$$

where τ_c is the yield stress = 11.063 Pa, K is the consistency = 33.785 Pas and n is the index = 0.424.

The Reynolds number for a non-Newtonian fluid is given by:

$$Re = \frac{\rho N d^2}{\mu_a} \qquad \textit{Eq. 3.10}$$

where d is the impeller diameter and ρ is the density.

From the previous equations, the Reynolds number was calculated:

$$Re = \frac{Nd^2\rho\dot{\gamma}}{\tau} = \frac{\rho N^2 d^2 k}{\tau_c + K(kN)^n} \quad \text{Eq. 3.11}$$

From this, the power number, N_p, of the mixer was estimated by linearly interpolating the results from Furukawa et al. using Plot Digitilizer (version 2.6.8) [143,147].

The power requirement of the mixer was calculated using:

$$W = \rho N_p N^3 d^5 \quad \text{Eq. 3.12}$$

An efficiency of 80% was assumed in order to calculate the overall power consumption of the mixing device.

3.4.4 Pump

Before entering the reactor, the feedstock was pumped to 24 bar in accordance with the pilot plant; this is roughly the saturation pressure of the products inside the reactor. The HTC reactor operates under autogenous pressure, that is the saturation pressure of water at the reaction temperature. A pressure drop of 3 bar was assumed inside the pre-heater double-pipe heat exchanger [135]. An efficiency, η, of 50% was assumed [148]. The work needed to raise the feedstock up to pressure (a total of 27 bar including the pressure drop) was calculated using:

$$W_{pump} = \frac{\Delta P \times \dot{V}_{feedstock}}{\eta} \quad \text{Eq. 3.13}$$

where ΔP is the pressure increase, $\dot{V}$ is the volumetric flowrate and η is the efficiency.

3.4.5 Reactor

The mass yields of the reaction products were estimated using experimental data and results of the statistical analysis as described in Chapter 4 HTC Test Programme. In order to calculate the size of the reactor, the volume of the feedstock in the reactor over the residence time was calculated. It was assumed that the reactor volume was 70% full and the size was determined accordingly [149]. The maximum volume of feedstock was set to 8 m^3 for each reactor in accordance with industrial design considerations. With a filled volume of 70%, this correlated to a maximum reactor size of 11.4 m^3. In cases where the volume of feedstock exceeded 8 m^3, multiple reactors were used. The ratio of height to diameter of the reactor was set as 3:7 as with the demonstration scale plant design. The internal pressure of the reactor was set as the vapour pressure of water at the reaction temperature.

HTC is an exothermic process; the heat of reaction for digestate was set as -0.75 MJ/kg_{feed} based on feedstock energy content, char energy content and combustion reactions [34]. Although this value was found at a temperature of 250 °C, it was assumed at all process conditions in the process model. Compared to traditional, dry thermochemical processes, the required energy to heat the water is small. The HTC system is closed and in saturated conditions and so the phase change from water to steam is avoided. Heat loss from the HTC reactor was modelled. The reactor was modelled as a pressure vessel, and therefore the reactor thickness was estimated using ASME Section VIII [150]. High-end values were used in this calculation for a conservative estimate: a pressure of 45 bar, temperature of 250 °C and diameter of 2 m, giving a reactor wall thickness of 49 mm. The insulation thickness was taken as 100 mm, the thickest insulation used in the demonstration scale plant design. All other values and constants used in heat loss calculations were taken from literature investigating the process energetics of HTC [122]. Overall, the heat loss from the reactor was calculated to represent less than 1% of the heat input to the feedstock and slurry mixture and so no further refinement was deemed necessary.

3.4.6 Depressurisation Tanks

HTC is carried out under autogenous pressure, up to 40 bar at 250 °C. Therefore, the mixture of hydrochar and process liquid, referred to as reaction slurry, exits the reactor via output pipes and is passed through a series of depressurisation tanks. There are 2 depressurisation tanks per reactor unit. Each depressurisation tank is connected to 2 outlet pipes. They are alternately filled and emptied from the reactor. The ratio of depressurisation tank height to diameter was set as 2:1 as per the demonstration plant design.

3.4.7 Ash-Reduction Equipment

The slurry from the depressurisation tanks then passes through a hydrocyclone for ash reduction. A reduction in the ash content was not modelled as experimental work would not give an accurate representation of industrial-size ash reduction. Therefore, the hydrocyclone was included for energy use and cost estimations only. The density of the slurry was recalculated using the moisture content and temperature of the mixture [141]. The electrical demand of the cyclone was estimated by scaling for the volumetric flowrate of the cyclone modelled by BMNT [22].

3.4.8 Filter Press

It was assumed that the filter press reduced the moisture content of the slurry down to 50 wt.% [151]. A temperature of 20 °C was used for the slurry, following cooling in the depressurisation and ash reduction equipment. The filter press was set to have up to 4 operations per day.

The volume of slurry produced per day was calculated assuming continuous operation of the reactor, equivalent to 7,800 hours/year. The pressure of the filter press was 8 bar, taken from the filter press design from the demonstration plant.

The work done by the filter press, W_{FP}, was calculated using:

$$W_{FP} = \frac{P_{FP} \times \rho_{FC}}{\dot{m}_{slurry}} \qquad \textit{Eq. 3.14}$$

where P_{FP} is the operating pressure of the filter press and ρ_{FC} is filter cake density at 20 °C.

The effective filter area of the filter press was calculated using a cross-sectional area of filter press membrane of 1.6 m^2 and a filter cake thickness of 0.04 m, both taken from plans for the filter press used at the demonstration plant. A maximum effective filter area was defined as 150 m^2 and multiple filter presses were modelled when the calculated area exceeded this [152].

The mass flowrate of filter cake was calculated using the defined moisture content of 50 wt.%. The mass flowrate of process water separated by the filter press was then calculated by subtracting the flowrate of filter cake from the flowrate of the slurry input.

3.4.9 Fluidised Bed Dryer

A fluidised bed dryer is used to dry the filter cake (moisture content of 50 wt.%) down to a moisture content of 12.5 wt.% as seen in industry, resulting in hydrochar. The fluidised bed dryer consists of a bed of filter cake and a mixture of hot process gases and air passing through at a high enough velocity to keep the filter cake in a fluidised state, resulting in high heat transfer and drying rates. The series of calculations used were taken from Volume 2, Coulson and Richardson [153].

The fluidising velocity, u_{fc}, was taken from Figure 16.26 in Coulson and Richardson [153] with the assumption that the mean particle diameter was 0.5 mm and the product particle specific gravity was 1.

The rate of evaporation, W, was calculated as:

$$W = \dot{m}_{HC,dry} \times (MC_{FC} - MC_{HC}) \quad \text{Eq. 3.15}$$

where MC_{FC} is the moisture content of the filter cake on a dry basis (kg/kg dry filter cake) and MC_{HC} is the moisture content of the hydrochar on a dry basis (kg/kg dry hydrochar).

Atmospheric air entered at a temperature (T_{in}) of 9.5 °C and the filter cake (T_{fc}) at 20 °C. The air was heated to a temperature (T_{air}) of 110 °C for the fluidised bed dryer. This temperature was similar to that as used in industry; it was limited as a higher temperature could lead to auto-combustion of the fine hydrochar particles. It was assumed that the bed temperature (T_b) and air exit temperature were both 70 °C meaning the latent heat of vaporisation (λ_b) of the moisture was taken as 2333 kJ/kg. The c_p of the air was determined by linear interpolation. The c_p of the filter cake was calculated based on the proximate analysis fractions and temperature [138]. The mass flowrate of air required was calculated by:

$$\dot{m}_{air} = \frac{\lambda_b W + c_{p,fc}\dot{m}_{fc}(T_b - T_{fc})}{c_{p,air}(T_{air} - T_b)} \quad \text{Eq. 3.16}$$

Therefore, the heat required to heat the air to this temperature was obtained by:

$$\dot{Q}_{FBD} = \dot{m}_{air} c_{p_{air}} (T_{in} - T_{air}) \quad \text{Eq. 3.17}$$

The diameter of the fluidised bed dryer (D) was then calculated by:

$$D = \sqrt{\frac{(\dot{m}_{air} + 1.58W)T_b}{278 u_{fc}}} \quad \text{Eq. 3.18}$$

A blower was used for the delivery of air to the dryer. The electrical energy associated with this use (W_{FBD}) was given by:

$$W_{FBD} = \frac{\dot{m}_{air} \times \Delta P_{FBD}}{\eta_{FBD}} \quad \text{Eq. 3.19}$$

The pressure difference across the dryer (ΔP_{FBD}) was set as 5 kPa [135]. The efficiency of the blower (η_{FBD}) was set as 75% [154].

3.4.10 Furnace

Although not part of the core HTC plant, a furnace following the fluidised bed dryer was added for instances where further thermal-treatment of the hydrochar was desired. This type of treatment has been shown to remove organic phytotoxic compounds and therefore could be useful for soil application of hydrochar [86,87]. A temperature of 275 °C was used as literature showed that this was the optimum temperature for eliminating phytotoxicity while maintaining the highest solid recovery of 73.5 wt.%. It was assumed that this thermal post-treatment densified the carbon content of the hydrochar by a factor of 1.06 based on a study using green waste hydrochar at similar conditions [87]. The resulting mass flow of treated hydrochar and its carbon content were calculated accordingly.

3.4.11 Heat Exchange Network

A heat recovery system was integrated in the plant using a series of heat exchangers and jacketed pipes containing water at 105 °C, maintained as a liquid under pressure. The system preheats the feedstock, recovers heat from the reaction products and preheats the air to the fluidised bed dryer. The heat requirement and possibility of heat recovery from the furnace was not included in the heat exchange network, giving a conservative estimate of the amount of heat recovery. The furnace was only included in the process model when called for.

The mass flowrate of water contained in the heat exchange network was scaled from the amount used in the demonstration plant. The potential for heat recovery was calculated and then any remaining heat demand was supplied by thermal oil heated by a gas-fired steam boiler. The heat recovery network is shown in Figure 3.5.

3.4.11.1 Feedstock Preheater

The feedstock was preheated before entry to the reactor using a jacketed pipe containing water from the heat recovery line. This heat exchanger was modelled as a counter-current double pipe heat exchanger with a minimum temperature difference of 10 °C, thus heating the feedstock to 95 °C. The overall heat transfer coefficient for this heat exchanger was set to 1504 W/m^2K (265 Btu/F ft^2 h) [154]. The remaining heat to raise the feedstock to reaction temperature was provided by the steam boiler.

3.4.11.2 Output Pipes Heat Recovery

Heat was recovered from the reaction products with a series of jacketed output pipes. There were 4 output pipes for each reactor. Again, these were modelled as double pipe heat

exchangers. Water from the heat recovery line is reheated to 105 °C in this section of the network. The same overall heat transfer coefficient as the Feedstock Preheater was used.

3.4.11.3 Air Preheater

The heat exchange network also preheated the air entering the fluidised bed dryer. As heat was exchanged between water and air, a larger minimum temperature difference of 30 °C was used, resulting in air preheated to 75 °C. All remaining heat to raise the temperature to 110 °C was supplied by the steam boiler and flue gas. The heat exchanger was modelled with a shell and tube design. A series of calculations were performed comparing the estimated lifetime cost of heat exchanger with larger surface area (higher CAPEX but lower OPEX from a reduced demand for natural gas supplying additional heating to the plant) or one with a smaller surface area (low CAPEX and high OPEX). These preliminary calculations found that a heat exchanger with a larger surface area had economic benefits over using a smaller heat exchanger over the assumed 20-year lifetime of the heat exchanger. The overall heat transfer coefficient was defined as 71 W/m^2K (12.5 Btu/F ft^2 h) [154].

3.4.12 Steam Boiler

Natural gas powered a steam boiler to supply the heat needed to raise the pre-heated feedstock to reaction temperature and the drying air used in the fluidised bed dryer to 110 °C. An efficiency of 85% was assumed for the steam boiler. The fluidised bed dryer is heated with a mixture of flue gas and pre-heated air. As the amount of flue gas was dictated by the amount of pre-heated air, the steam boiler duty and subsequent size were calculated iteratively until an error of 0.05% was achieved between the duty required and supplied.

3.5 Capital Expenditure

Capital cost (CAPEX) estimation tools from three book sources were used to calculate a preliminary capital cost of the HTC plant with an accuracy of ±30%. The factorial method detailed in Seider et al. was followed [154]. The CAPEX of the plant was validated with confidential data for a demonstration-scale HTC facility. Due to differences in the labelling of plant equipment, each unit was categorised into feeding equipment (upstream of the HTC reactor), the reactor, separation (downstream of the HTC reactor), heat exchangers and boiler for comparison.

Equations from Table 16.32 of Seider et al. [154], Table 7.2 of Towler and Sinnott [155], and Appendix D from Woods [152] were used to calculate the free on board (f.o.b.) purchase cost of the plant equipment. All purchased cost equations assume that the unit is made of carbon steel. However, the majority of the plant equipment was stainless steel and therefore a

material factor was used to convert the cost. Woods [152] gives a specific material factor for each type of plant equipment and these were used where applicable. When equations from Seider et al. [154] were used, a basic materials factor of 2 for stainless steel was used. References and size factors for each piece of equipment are shown in Table 3.4. Also reported in are the maximum equipment sizes. As described in Section 3.4 Process Model, the industrial plant follows a modular design. Based on the industrial designs, the maximum feedstock contained by the reactor in this model was defined as 8 m^3, which correlated to a maximum reactor volume of 11.4 m^3. The maximum size of the rest of the plant equipment was based on the upper limits of the CAPEX estimations in literature. Whenever the size of the plant equipment exceeded this maximum, multiple units were used.

*Table 3.4: Equipment type and size factors used for estimating the f.o.b. purchase cost of the plant equipment. *An industrial quote was used, scaled to size using the scaling factor by Woods* [152].

Operation	Equipment type	Size factor	Maximum size	Ref
Biomass Hopper	Vibratory feeder, solid-handling system	Volumetric flowrate	900 ft^3/h = 0.007 m^3/s	[154]
Mixer	Propellers, top entry, open tank	Drive power	75 kW	[152]
Feedstock Pump	Pumps, reciprocating, piston or plunger type	Drive power	200 kW	[152]
Feedstock Preheater	Double pipe heat exchanger	Area	80 m^3	[155]
Reactor	Pressure vessel, horizontal or vertical	Volume	11.4 m^3 (industrial)	[152]
Output Pipes	Double pipe heat exchanger	Area	20 m^3	[152]
Depressurisation Tanks	Pressure vessel, horizontal or vertical	Height x $diameter^{1.5}$	0.7 m^3	[152]
Hydrocyclone	Hydrocyclones	Liquid feed rate	1,200 gal/min = 0.0013 m^3/s	[154]
Filter Press	Filter, plate and frame	Effective filter area	150 m^2	[152]
Air Preheater	Shell and tube heat exchanger	Area	2,000 m^3	[152]
Fluidised Bed Dryer*	Industrial quote for fluidised bed dryer	Diameter	10 m	[152]
Furnace	Furnaces, cylindrical	Duty	60 MW	[155]
Steam Boiler	Steam boiler	Heat absorbed	70 MBtu/hr = 20.5 MW	[154]

The capital cost estimates were initially calculated based on the US Gulf Coast. They were scaled using the average cost index (CEPCI) in 2019, converting from USD to GBP and applying the UK location factor. Details of these are shown in Table 3.5.

Table 3.5: Cost conversion factors.

Index	2019 Average	Ref
Chemical Engineering Plant Cost Index (CEPCI)	607.5	[156]
USD to GBP conversion	0.784	[157]
EUR to GBP conversion	0.877	[158]
Harmonised Index of Consumer Prices (HICP)	107.8	[159]
UK location factor	1.05	[160]

The free on board (f.o.b.) purchase cost only accounts for the equipment itself and not the costs of the auxiliary equipment or installation. For some of the operating expenditure (OPEX) calculations, estimates of the inside battery limits (ISBL) and offsite costs were needed. Towler and Sinnott presented factors to calculate these from the f.o.b. cost [155]. To scale the purchased cost to the installed cost, the modified Lang factor method found in Seider et al. was used [154]. Seider et al. provides factors for three types of processes: fluids, fluids-solids and solids. The fixed capital expenditure is what is commonly known as the CAPEX and represents the cost of the plant equipment. The total capital investment includes the working capital, this is what is required to start up the plant and keep it running. When calculating the fixed capital investment or total capital investment, a factor of 1.05 was included to account for the delivery of equipment to plant site. These factors are summarised in Table 3.6.

Table 3.6: Factors used to convert purchase costs to fixed capital investment.

Factor	Process type			Ref
	Solids	Solids-fluids	Fluids	
ISBL	2.5	3.2	3.3	[155]
Offsites	0.4 × ISBL	0.4 × ISBL	0.3 × ISBL	[155]
Fixed capital investment (CAPEX)	3.97	4.28	5.04	[154]
Total capital investment (Lang factor)	4.67	5.03	5.93	[154]

3.6 Operating Expenditure

The operating cost (OPEX) was divided into two categories: variable production costs and fixed production costs. The methodology was mostly taken from two book sources [161,162]. Both variable and fixed operating costs were reported on an annual basis.

3.6.1 Variable Production Costs

Apart from the dewatering cost calculations, no consumables costs were considered as their use was minimal. Dewatering consumables were included in the dewatering calculations. Consumables refer to solvents, acids, bases, inert materials, corrosion inhibitors, additives, catalysts and adsorbents.

Natural gas was used to provide all additional heat not supplied by the heat recovery network. The cost of natural gas in the UK was taken as the 2019 average for a small-medium sized consumer (500–1,999 MWh annual consumption) given by the Department for Business, Energy and Industrial Strategy, 2.255 p/kWh [163]. This was then converted to £/J and the annual natural gas cost was calculated assuming 7,800 hours of operation a year.

The unit operations using electricity were the mixer, pump, hydrocyclone, filter press and fluidised bed dryer. The cost of electricity in the UK was taken as the 2019 average for a medium sized consumer (2,778–27,777 MWh annual consumption) given by the Department for Business, Energy and Industrial Strategy, 12.78 p/kWh [163]. This was then converted to £/J and the annual electricity cost was calculated assuming 7,800 hours of operation a year.

The only waste considered in this analysis was the process water, which was directed to wastewater treatment. A cost of $1.5/tonne was used for the wastewater treatment cost [155]. Directing process water to wastewater treatment was used as the default condition as there are expected complications with its use on land, such as the nitrogen limits set out in the PAS110 specifications for organic fertilisers [164]. However, process water has been shown to contain valuable concentrations of nutrients at certain process conditions and so its use as a fertiliser has been assessed in Chapter 4 HTC Test Programme and Chapter 6 Case Study. Any ash removed using the cyclone technologies was not considered as the removal rate was not calculated.

3.6.1.1 GHG emissions

The greenhouse gas (GHG) emissions from the use of natural gas and electricity in the HTC plant were estimated using conversion factors from 2019 [5]. The Department for Business, Energy & Industrial Strategy reported emissions of 0.1839 kg CO2e/kWh for natural gas (gross) and 0.2556 kg CO2e/kWh for electricity based on the average UK electricity mix. The transmission and distribution loss factor for UK electricity was 0.0217 kg CO_2e/kWh, bringing the total GHG emissions related to the UK electricity mix to 0.2773 kg CO_2e/kWh.

However, if biogas from the AD plant producing digestate was used as fuel, the emissions would reduce to 0.00021 kg CO2e/kWh, based on a biogas methane content of 65%. These

calculations do not account for GHG emissions from transportation, dewatering, distribution or any other parts of the process.

3.6.2 Fixed Production Costs

In contrast to the variable production costs, fixed operating costs do not depend on the plant's running capacity. The fixed production costs were labour, supervision and management, direct salary overhead, maintenance, land, insurance, interest, admin, research and development, and distribution and selling (Table 3.7). Unless stated otherwise, the methodology was taken from Towler and Sinnott [162].

*Table 3.7: Fixed production cost sources. *The number of shift positions was taken from the industrial HTC business plan.*

Operation	Details	Ref
Operating Labour*	Average annual pay for operator = £23,000 Number of shift positions (#SP) depends on the number of reactors (#R): #R: 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11+ #SP: 2 | 2 | 3 | 4 | 5 | 5 | 6 | 6 | 7 | 7 | 8+ 4.8 operators per shift	[162,165]
Supervision and management	25% of operating labour	[162]
Direct salary overhead	40–60% of labour plus supervision costs (50% was used)	[162]
Maintenance	3–5% of ISBL (4% was used)	[162]
Land	1% of ISBL + OSBL	[162]
Insurance	1% of ISBL + OSBL	[162]
Interest Payments	3.7% the working capital, the median interest rate charged to SMEs in the UK for the credit line or bank overdraft for which they applied	[166]
Administrative costs	25% of direct salary overhead	[161]
Research and development	5% of the total expenses	[161]
Distribution and selling	10% of the total expenses	[161]

3.7 Market Analysis

HTC of wet, waste biomass lends itself to two primary revenue streams: sale of the HTC products and savings in comparison to current waste treatment technologies. Both of these are investigated here, reporting the sale prices of different product markets and an estimate of the cost of current digestate treatment. Ultimately, these will be used alongside the

predicted costs of HTC to perform an investment analysis and assess the financial viability of HTC for the treatment of digestate.

3.7.1 HTC product market

The different product markets for the HTC products were assessed using a combination of commercial, literature and governmental sources. All costs were converted to GBP in the year 2019 using the cost conversion factors shown in Table 3.5. The sale prices of the most common HTC products and their markets are shown in Table 3.8.

Table 3.8: Sale prices and comparative costs for a range of HTC products.

Product type	Market	Sale price	Source
Hydrochar	Solid fuel	£122–£188/tonne £8.46/GJ	NEWAPP project, focussed on the industrial application of HTC [136]
Hydrochar, process liquid	Fertiliser	£0.61/kg N £1.44/kg P £0.56/kg K	Converted from GB fertiliser prices given by the Agriculture and Horticulture Development Board [167]
(Treated) hydrochar	Carbon sequestration	£418/tonne C	Based on the cost of abatement of the HyNet North West England Project [168]

Certain product characteristics must be met for each of these different applications. For hydrochar as a solid fuel, a high energy content, usually indicated by a high carbon content, is valuable. In addition to this, the ash content must be kept to a minimum and a high ash melting temperature is desirable to reduce the effect of slagging and fouling in the boiler. Digestate feedstocks tend to be higher in ash than lignocellulosic biomass and so the market potential of hydrochar from digestate is examined.

For soil application, either process liquid as a fertiliser or hydrochar as a means of carbon sequestration, nutrient and heavy metal concentration limits must be adhered to. The PAS110 legislation specifies limits for digestate and compost spreading to land [164]. Of particular concern for digestate and its products is the risk of exceeding the nitrogen application limits in Nitrate Vulnerable Zones (NVZs). Storage requirements resulting from this can come at a high capital cost to the operator [23]. On top of this, there are concerns about the growing area of land designated as a NVZ in the UK. Draft legislation in Wales from April 2020 proposed to designate the whole of Wales as a NVZ, over 40 times the area currently covered [169]. There are existing standards concerning trace elements and heavy metal concentrations for biochar and hydrochar products. The European Biochar Certificate (EBC) was created in 2013 to standardise the use of biochar in agriculture [92]. The Biochar Quality Mandate (BQM) is a similar set of standards developed in the UK [170]. Therefore, the HTC products have been

compared against these to assess their suitability for land application. Fuller explanations and considerations are given in Chapter 4 HTC Test Programme and Chapter 6 Case Study.

3.7.2 Feedstock Market

The anaerobic digestion market in the UK is growing, with new applications and facilities under development each year [19]. The AD Portal Biogas Map , WRAP Organics recycling industry status report, and ADBA AD map were used to estimate and predict digestate production in the UK [15,16,171]. It is expected that an increase in food waste treatment will be a contributing factor to a rise in digestate production in the coming years, and forecasts from the WRAP report Food Surplus and Waste in the UK were used in this analysis [172].

In order to investigate the financial viability of the HTC of digestate, the cost of current digestate treatment was calculated. The standard practice for managing digestate is land spreading, with the criteria for this set out in the PAS110 [164]. Details of the cost of transporting and spreading were used to make an estimate [20,21]. Due to the high nitrogen content of digestate, there is further legislation relating to its storage and spreading [173]. The additional resulting cost of this was used in analysis [22,23].

Aside from comparing to standard practice, the cost of transporting wet feedstock to the HTC plant was incorporated in the transport model. Two English counties were used as case studies for this: Lincolnshire as the largest producer of digestate in the country, and Nottinghamshire as the largest digestate producer relative to the county's size. It was assumed that the whole digestate was dewatered down to a moisture content of 75 wt.% before being transported to the HTC plant.

Data for all the AD facilities in both counties was collected from the AD Biogas Map [15]. The counties were split into a large grid with each interval representing 100 m^2. The cost of transporting all the digestate in the county to each interval was calculated using an estimate for the transportation of digestate fibre [21]. The interval that gave the overall lowest cost for all the digestate fibre in the county was deemed to be the optimum location for the HTC plant. Once the postcodes of the optimum HTC plant locations had been found, driving distances from each of the AD plants in the county were then measured to gain a more accurate transportation distance and thus transportation cost. The plants were ordered in distance from the optimum location and a cumulative sum of digestate fibre to be transported to the plant and the transportation cost associated with that were calculated. A second-order polynomial relationship was found for each of the counties relating total transport cost and plant capacity.

3.8 Conclusions

Three types of digestate were used experimentally, produced by the AD of different feedstocks: waste-based, farm-based and crop-based. These were selected to represent the most common forms of digestate in the UK. Multiple HTC runs were performed for each feedstock with varying conditions of temperature (180–250 °C), residence time (60–180 minutes) and moisture content (80–95 wt.%). These reflected the HTC conditions commonly seen in literature and industry [137]. The mass yields of each of the products were measured following experimentation.

The elemental and proximate content of the digestate feedstocks and HTC products were measured using a CHN analyser and TGA. Hydrochar samples from another food-waste based digestate were also sent to be tested in germination and plant growth trials.

Regression models were created using the experimental results in order to predict the mass yield and HTC product properties. These were then fed into a process model of an industrial-scale HTC plant. The process model was designed using industrial data for an existing HTC plant. Cost models from literature and publicly available data were used to estimate the capital and operating costs in the process model.

Forecasted HTC product sale prices and savings over existing digestate treatments were then used to perform a market analysis. The economic feasibility of HTC as a digestate treatment could then be assessed using an investment analysis and market predictions.

4 HTC Test Programme

4.1 Introduction

The aim of this work was to investigate how three different types of digestate feedstock performed in HTC at different process conditions and to explore how well the results could be modelled. To meet these aims, the following work is presented:

- Results of the hydrochar mass yield and carbon contents for each of the feedstocks at different processing conditions.
- The fate of organic and inorganic compounds in the different HTC product fractions.
- An assessment of the suitability of hydrochar and process liquid for solid fuel, fertiliser, and soil amendment markets.
- An overview of the most important factors in the regression models along with a more in-depth study of the hydrochar yield on a dry ash-free basis, the hydrochar carbon content on a dry ash-free basis, and the hydrochar ash content.
- A comparison of the regression model developed in this work with those found in literature.

Results of the regression models are used later in the HTC process modelling work. While only a selection of the experimental results are reported, the full data set is included in the Appendix, Table A.1 and Table A.2.

4.2 Feedstock

Hydrothermal carbonisation was carried out in a 75 ml reactor for each of the feedstocks under a range of different conditions. Details of the experimental HTC procedure and the feedstocks used are given in Chapter 3 Methodology. Table 4.1 summarises the HTC runs carried out for each of the feedstocks. Experimental results for these runs can be found in the Appendix, Table A.1 and A.2. As discussed in Chapter 3 Methodology Section 3.3 Statistical Analysis of Results, a full Box-Behnken design of experiments was performed for the waste-based digestate. This design was chosen as similar HTC experimental studies have produced regression models with good fit [40,43]. However, access to the farm-based and crop-based digestate samples was restricted and therefore only 6 runs were performed rather than the 15 required for a 3-factor Box-Behnken design of experiments. Better predictive capabilities were expected from the regression equations derived from the waste-based digestate due to the more comprehensive experimental design. Temperature was shown to be the factor with by far the largest influence on HTC, as found in literature [5,41,50–53,42–49]. By investigating the effect of temperature for the farm-based and crop-based digestates, it was anticipated that any shortcomings from performing a limited number of experimental runs were minimised.

Table 4.1: Process conditions for the HTC runs carried out for each feedstock.

	Waste-based	Farm-based	Crop-based
Number of runs	15 (Box-Behnken design of experiments)	6	6
Temperature	180–250 °C	190–250 °C	180–250 °C
Residence time	60–180 minutes	60–180 minutes	60 minutes
Moisture content	80–95 wt.%	85 wt.%	75–95 wt.%

Each of the digestates were dried at 105 °C for 24 hours and then characterised. Their compositions are shown in Table 4.2. Details of the proximate and ultimate analysis procedures are included in Chapter 3 Methodology, Section 3.2.3 Sample Analysis.

*Table 4.2: Digestate feedstock properties. VM is volatile matter and FC is fixed carbon. Values in brackets are on a dry ash-free (daf) basis. *Oxygen content was calculated by difference.*

		Waste-based	Farm-based	Crop-based
Proximate analysis	VM (daf wt.%)	93.9	77.4	77.7
	FC (daf wt.%)	6.1	22.6	22.3
	Ash (dry wt.%)	36.5	22.9	9.3
Ultimate analysis (wt.% dry) (daf in brackets)	Carbon	27.6 (43.5)	38.6 (50.1)	39.9 (44.0)
	Hydrogen	3.8 (6.0)	4.5 (5.9)	5.0 (5.5)
	Nitrogen	3.5 (5.5)	3.7 (4.8)	2.3 (2.5)
	Oxygen*	29.5 (46.4)	33.7 (43.7)	46.6 (51.4)

AD plants can accept and process a wide range of substrates, and the digestate feedstocks used experimentally were selected to encompass these. They were categorised as waste-based, farm-based and crop-based and compared with digestate characteristics from literature to explore the representativeness of the samples. Figure 4.1 shows the digestates grouped into categories and their ash, carbon, hydrogen, nitrogen oxygen contents. While there are noticeable variations within each category, there are visible trends. The ash content tended to be higher in the waste-based digestate followed by the farm-based digestate and then the crop-based digestate. The carbon content was lowest for the waste-based digestate, but similar for the farm-based and crop-based samples. While the chosen samples are representative of their categories, relatively small samples were tested and the heterogenous nature of digestate is also worth consideration. An AD plant processing a particular crop will generally produce digestate with consistent characteristics. From observing of the AD Portal Biogas Map and discussions with farm-based AD operators, it is understood that farm-based

AD plants tend to process a wider range substates. This is often governed by seasonal changes in substrate availability and results in varying digestate composition throughout the year [15]. Waste-based digestate is also expected to be more variable in nature due to the heterogenous nature of waste itself. General conclusions can be drawn up from the experimental findings from these digestate samples, but some variation would be expected if the samples were taken from the same plants at a different point in the year. Figure 4.2 shows photos of the digestates used at different stages of their processing in the laboratory.

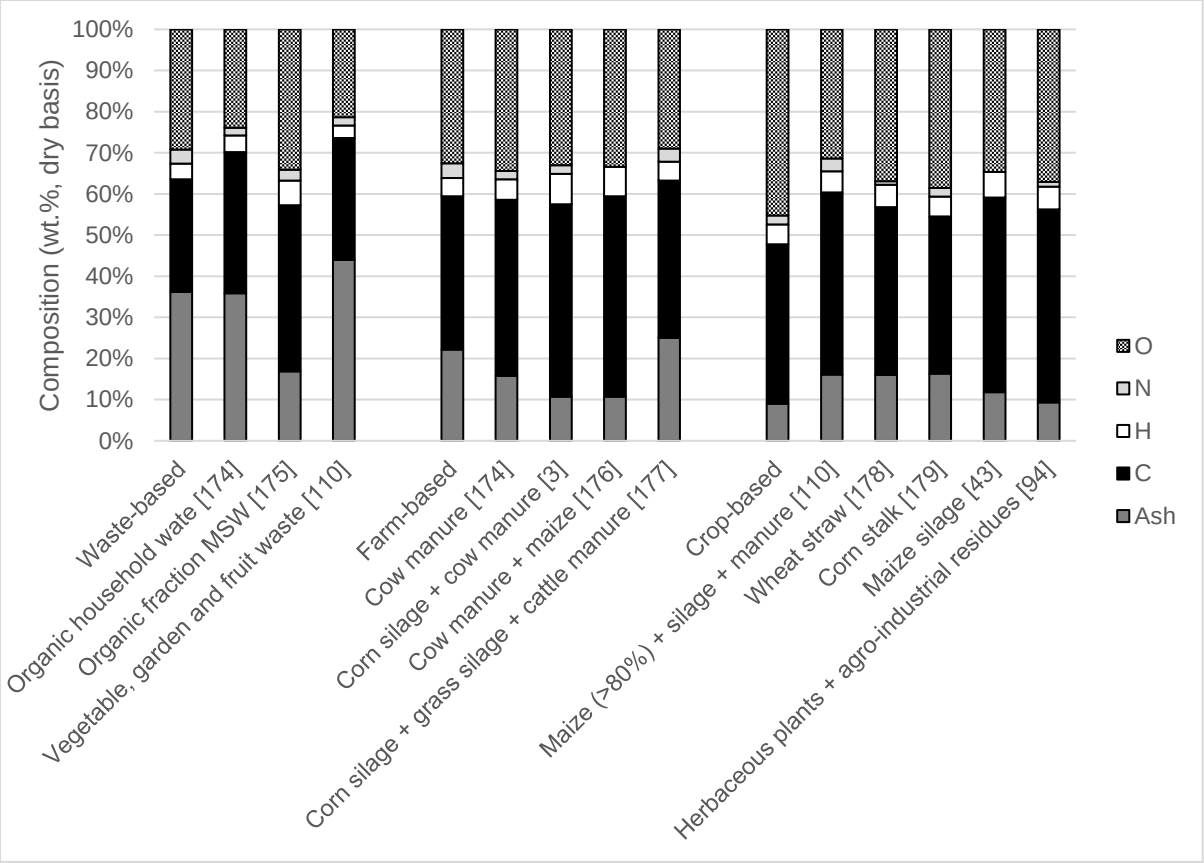


Figure 4.1: Composition of digestate samples used in this work compared to literature results.

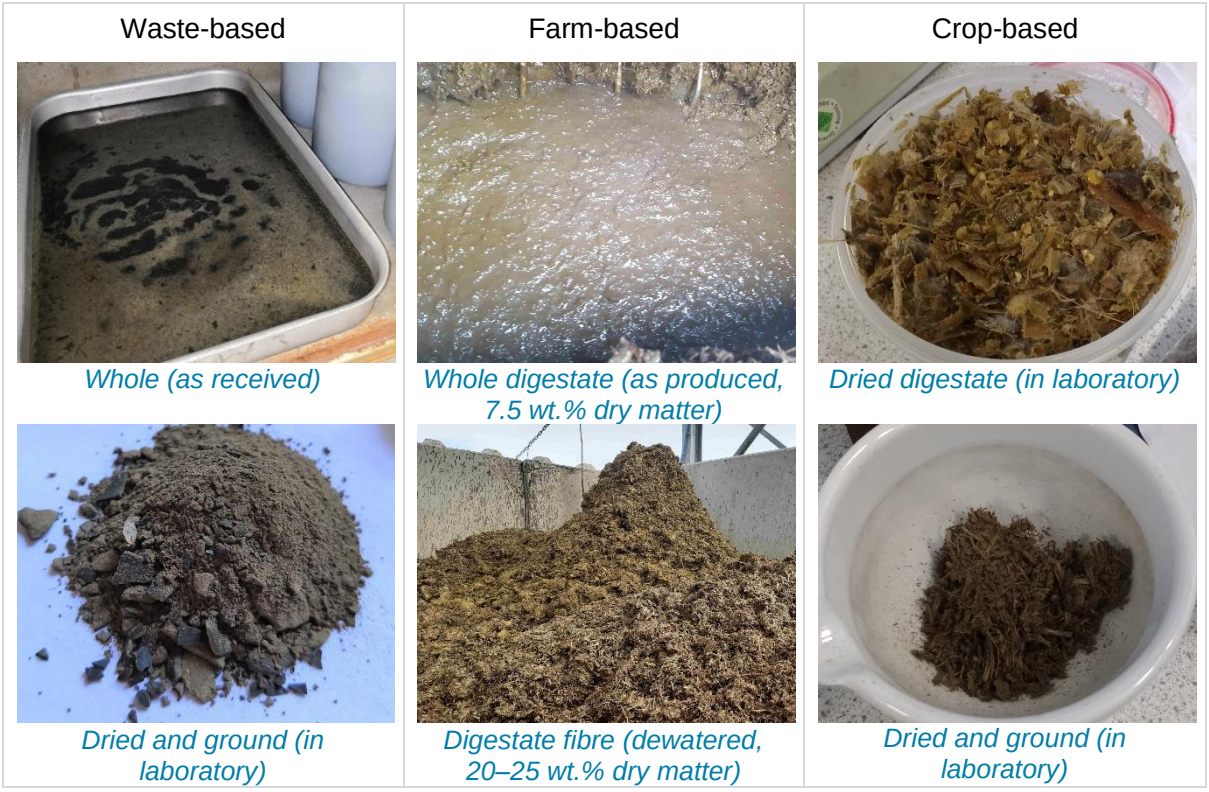


Figure 4.2: Photos of the digestates used experimentally at different stages of feedstock preparation.

4.3 Experimental results

4.3.1 Mass yield and carbon content

At first glance, it was clear that HTC temperature had the largest influence on the hydrochar yield and properties, as found in literature [5,41,43]. A more detailed consideration of the effect of HTC severity is given later in Section 4.4 Regression model. Figure 4.3 shows the hydrochar and process liquid produced from crop-based digestate at 180 °C and 250 °C. It can be seen that at lower HTC temperatures, the hydrochar retained some of the original feedstock texture and was produced with a higher yield. Conversely, higher HTC temperatures resulted in hydrochar with finer particles but a higher quantity of process liquid.

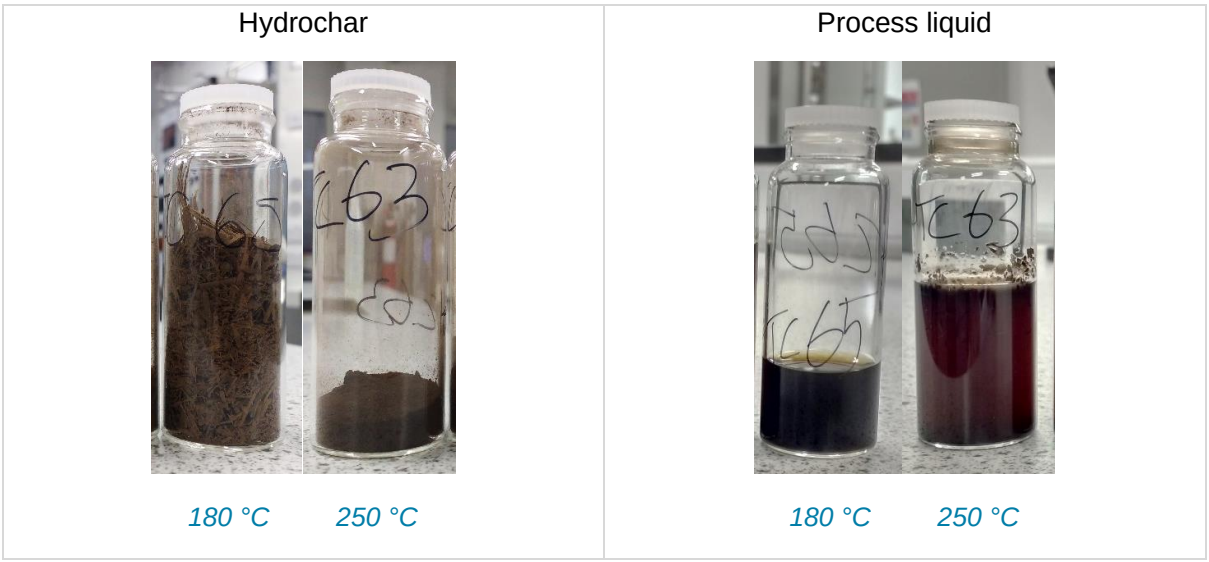


Figure 4.3: Photos of hydrochar and process liquid from the crop-based digestate with a residence time of 60 mins and a biomass moisture content of 89 wt.% at 180 °C and 250 °C.

Figure 4.4 shows a negative trend between HTC temperature and hydrochar yield for each of the feedstocks. Conversely, the yield of process liquid increased with an increase in HTC temperature. The yield of process gas was relatively low for all experimental runs, averaging 4.7 wt.%. At lower temperatures, the solid mass yield of the crop-based digestate was higher than for the other two feedstocks. At higher HTC temperatures, the difference in hydrochar yields for the feedstocks narrowed.

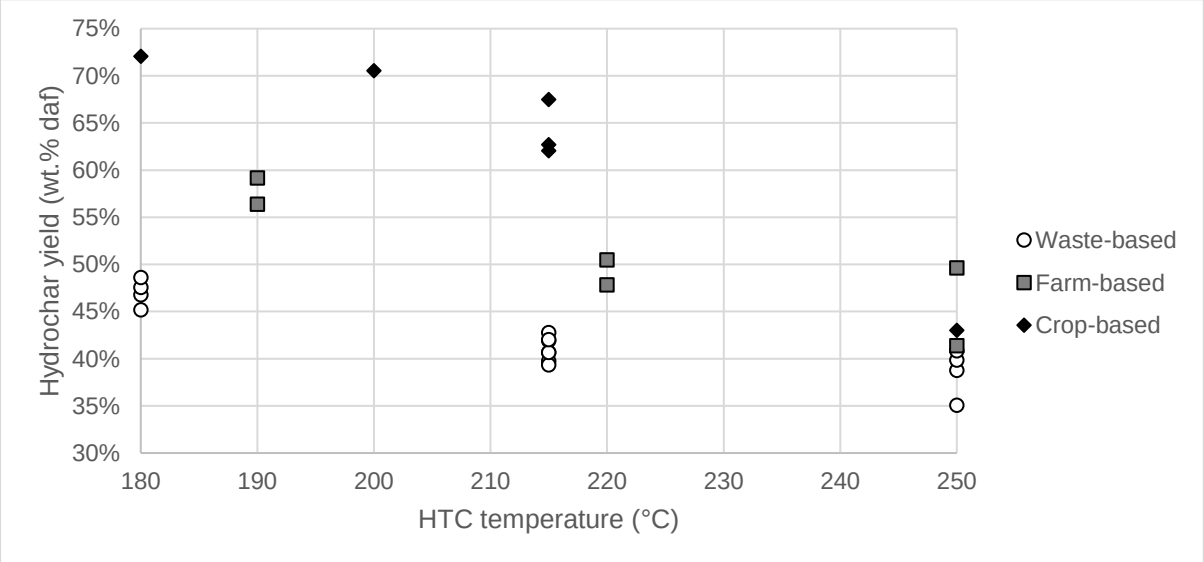


Figure 4.4: Effect of HTC temperature on the hydrochar yield for each of the digestate feedstocks. The dry ash-free (daf) yields were calculated as a percentage of the daf mass of the feedstock used.

A positive trend was seen for hydrochar carbon content and increasing HTC processing temperature for all three types of digestate, as shown in Figure 4.5. The hydrochars produced from the farm-based digestates had the highest carbon contents on a dry ash-free basis.

However, due to the much lower ash content of the crop-based digestate, it was the crop-based hydrochar that had the highest carbon content overall on an as-received, dry basis.

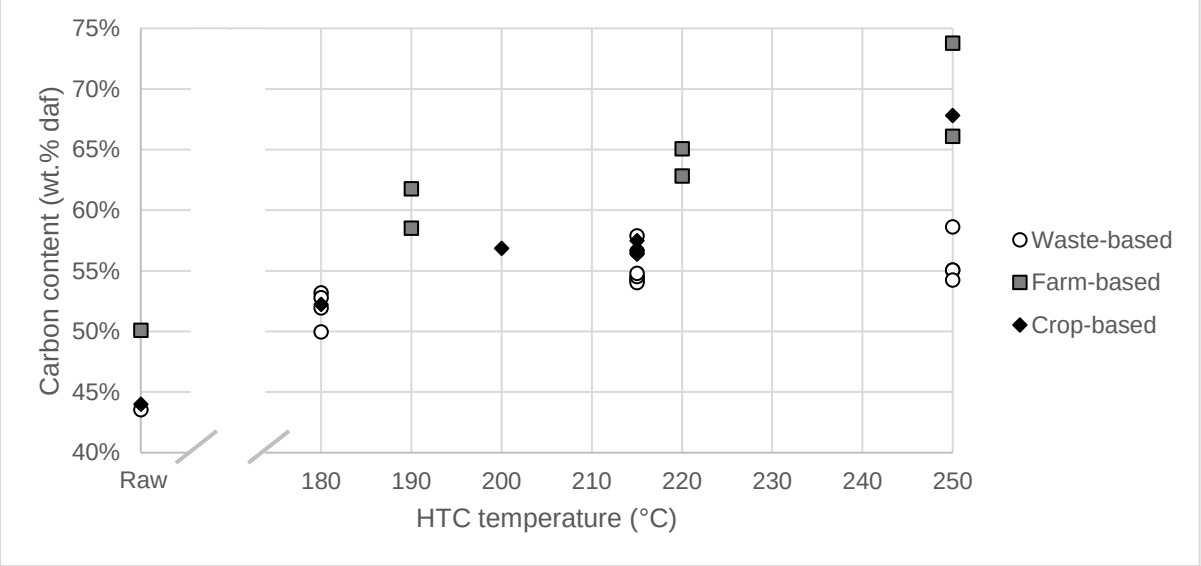


Figure 4.5: Effect of HTC temperature on the hydrochar carbon content (dry, ash-free) for each of the digestate feedstocks.

Generally, an increase in HTC temperature led to an increase in the degree of carbonisation of the hydrochar; that is, the hydrochar had a higher carbon content but a lower mass yield. This was observed for all three of the digestate samples and is supported by literature findings [30]. This trade-off between hydrochar carbon content and mass yield is an important consideration for HTC operators, and is explored further in Chapter 5 Process Model.

4.3.2 Elemental balance

Carbon, hydrogen and nitrogen concentrations of the digestate, hydrochar and process liquid samples were measured using elemental analysis. The oxygen concentration was calculated by difference. The waste-based digestate was used to study the carbon and nitrogen balance. Figure 4.6 shows the recovery in the solid hydrochar, process liquid and process gas fractions of the HTC products. Each sample was run in triplicate and the percentage recovery from the original digestate feedstock is shown. Each bar represents a different set of HTC process conditions in ascending temperature; the quantities of process liquid recovered from 2 of the HTC runs were not large enough for the CHN analyser and so results are given for only 13 of the 15 runs. It was deduced that for the waste-based digestate, an increase in HTC temperature led to a slight decrease of both carbon and nitrogen recovery in the hydrochar. The dependence of carbon recovery on HTC temperature is explored further using regression models and also in Figure 4.18 (Section 4.4.2 Model Results). Figure 4.6 shows that overall carbon recovery varied between 63% and 106%, with an average of 86%. For the nitrogen,

the recovery varied between an apparent 57% and 140% with an average of 106%. For both carbon and nitrogen readings, it is likely that there was a higher degree of error in the liquid analysis given the small variation expected for the hydrochars. From the results, the carbon recovery results gave an underestimate, but many results fell within a range of ±20%. The nitrogen results, however, varied much more. One reason for this could be the loss of ammonia during the preparation of the digestate and hydrochar samples which lead to an artificially high nitrogen recovery measured in the process liquid. Previous work has suggested that hydrochar is able to absorb ammonium well due to its acidic nature, but this sorbed ammonia could then be lost during post-treatment, either through washing or drying at 105 °C [180]. The waste-based digestate used experimentally was dried at 105 °C and had a measured nitrogen content of 35,000 ppm on a dry basis. However, independent testing of this same wet feedstock, used as part of the PAS110 certification for digestate, gave the nitrogen content as 90,000 ppm on a dry basis, with an ammoniacal nitrogen content of 72,000 ppm. Therefore, it seems likely that loss of ammonia occurred during the sample preparation of digestate prior to testing and equally so with the hydrochar. However, it is important to note that digestate is a heterogenous feedstock and there are changes in composition seasonally depending on what is input to the AD unit. Therefore, a contributing factor to the difference in composition between the laboratory sample and raw sample could be due to the variable quality of digestate itself.

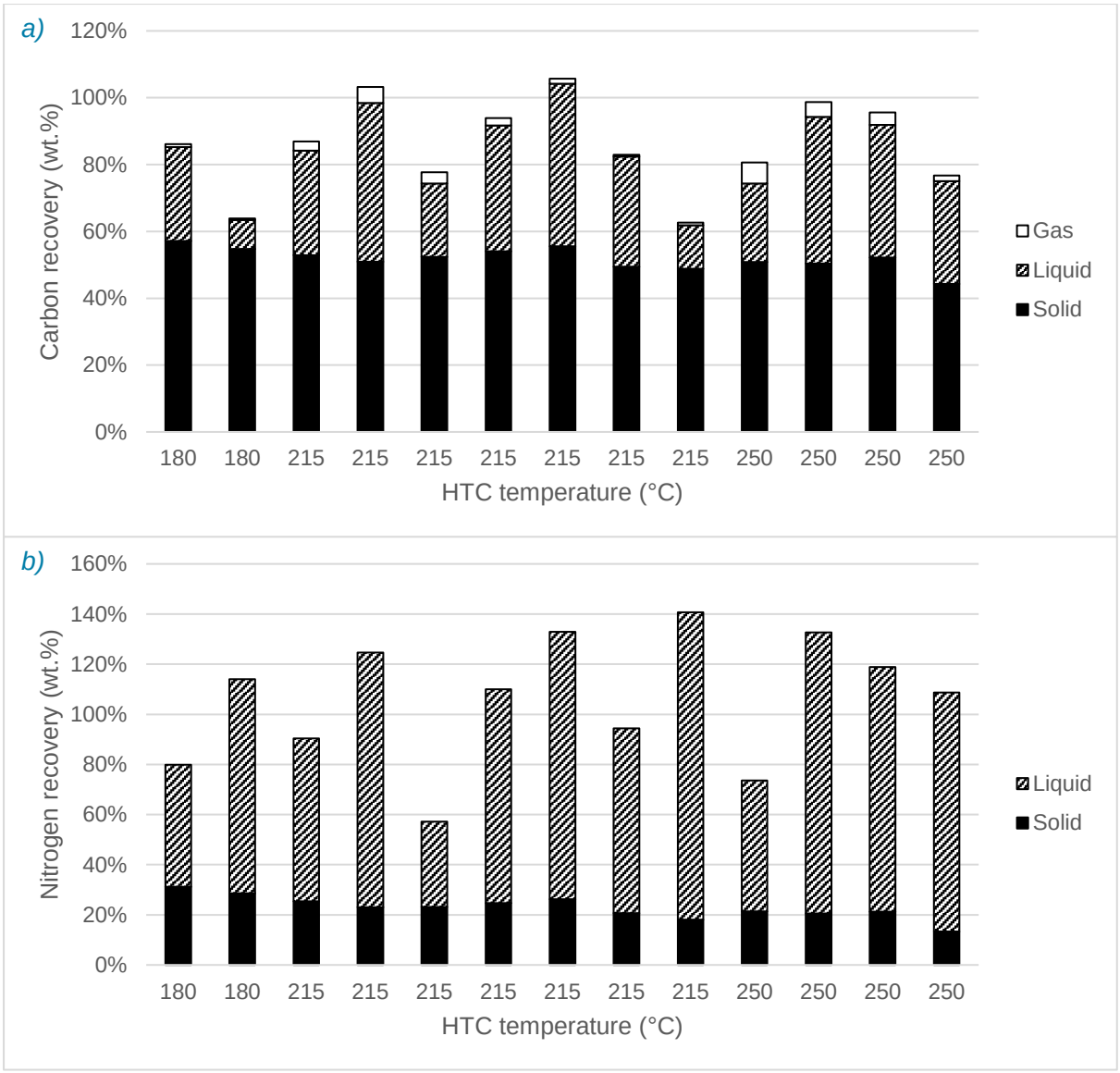


Figure 4.6: a) carbon and b) nitrogen recovery in the solid hydrochar, process liquid and process gas (assumed 100% CO_2) from the HTC of waste-based digestate at 13 different sets of reaction conditions.

4.3.3 Fate of inorganic elements

The inorganic elemental content of the farm-based and waste-based digestate and process liquid samples were measured. The inorganic element content of the hydrochar samples were calculated by difference. These digestates had much higher ash contents than the crop-based digestate and so their ash content and the potential use of HTC products for fertiliser or soil amendment was of greater interest. Figure 4.7 shows the ash composition of the waste-based and farm-based digestates.

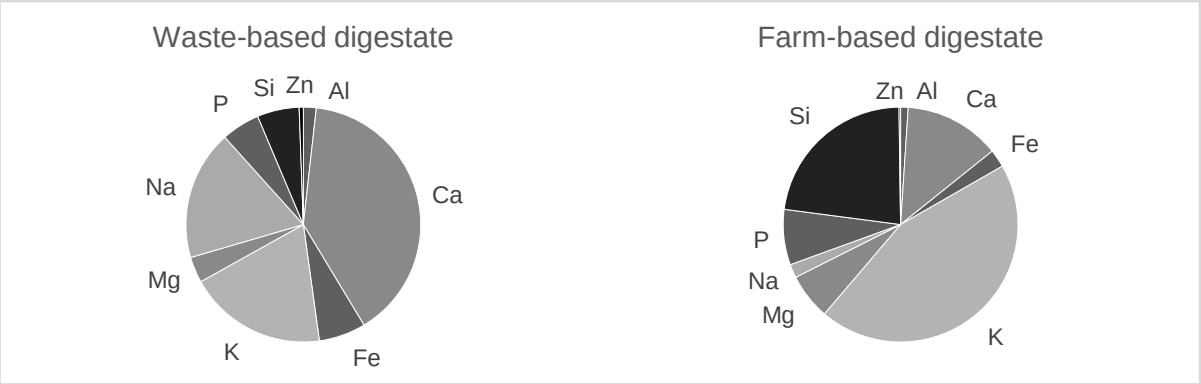


Figure 4.7: Inorganic composition of the ash of the waste-based and farm-based digestates.

Figure 4.8 shows the fate of inorganic elements. These results are for HTC runs at 215 °C, 180 minutes and an 80 wt.% moisture content for the waste-based digestate, and 220 °C, 180 minutes and an 85 wt.% moisture content for the farm-based digestate. These conditions were selected as they were the mid-range temperatures and most similar to each other for comparison. Aside from potassium and sodium, the inorganic elements almost wholly remained in the solid hydrochar after HTC. This trend is consistent with Smith et al. [73] who found potassium and sodium recovery in hydrochar to be the lowest of the elements they studied. As both phosphorus and potassium were of interest when considering the fertiliser potential of the digestate, hydrochar and process liquid, their concentration as a function of the process severity was studied with details given in Section 4.3.4.2 Fertiliser Potential.

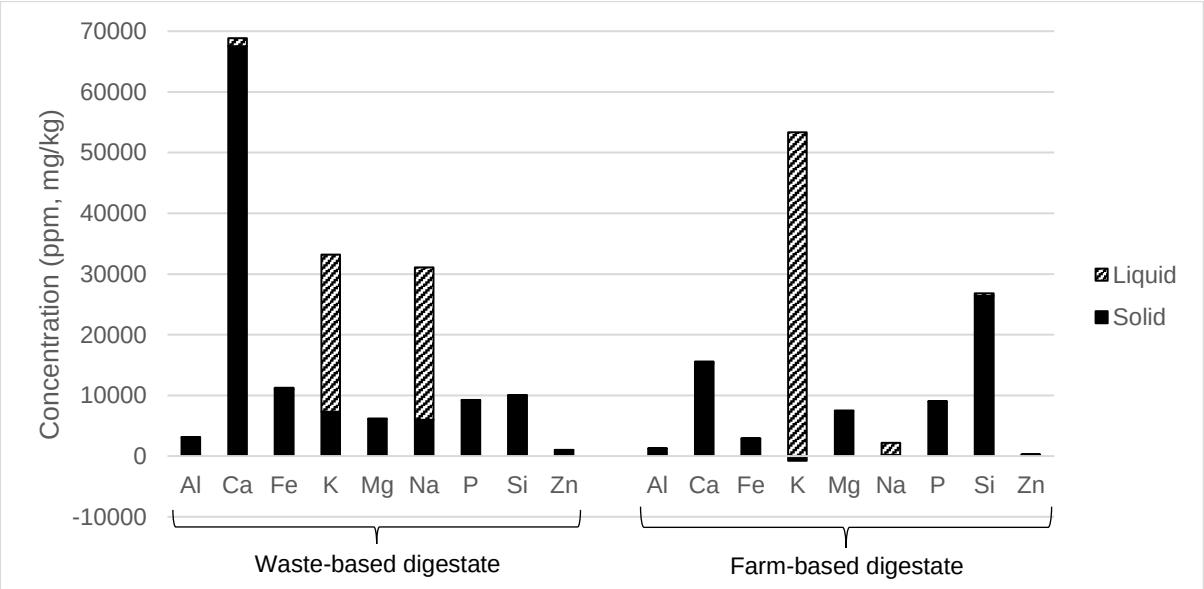


Figure 4.8: Concentration of inorganic elements in the solid hydrochar and process liquid. Process liquid concentrations were measured using ICP analysis and the hydrochar contents were calculated by difference from the original feedstock.

The recoveries in hydrochar in Figure 4.8 were calculated by difference. This explains why a negative recovery was found for K for the farm-based digestate. In order to investigate the

validity of calculating the recovery by difference, ICP analysis was also performed on two hydrochar samples from the farm-based digestate. Figure 4.9 shows the average percentage recovery of the inorganic elements measured in the farm-based digestate. The left-hand bar represents HTC at 190 °C and the right-hand bar is for products formed at 250 °C. There are differences in recoveries at these two different temperatures but with only two data points, they were not considered comprehensive enough to draw up any conclusions about the effect of temperature. All these readings fall within a range of 100±15%, apart from calcium which gave an apparent recovery of 133%. Phosphorus and potassium, of high importance when considering fertiliser use, balanced well. Phosphorus readings seemed to give a slight under-estimate, of down to 92%, and potassium results gave a slight over-estimate of up to 112%. It was concluded that the calculation of inorganic content in the hydrochar by difference was generally valid but testing the hydrochar separately is recommended when considering the fertiliser potential of hydrochar itself.

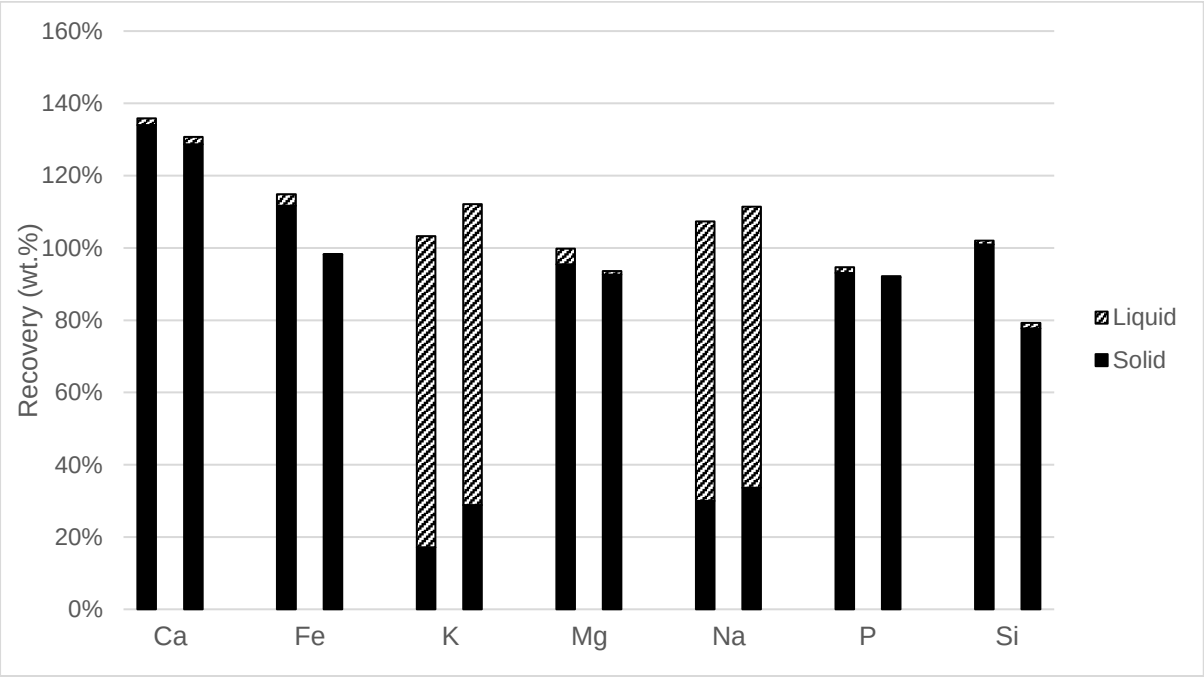


Figure 4.9: Percentage recovery of major elements in the inorganic content of the farm-based digestate, calculated using the process liquid and hydrochar ICP results. The left-hand bar represents a HTC temperature of 190 °C for 180 minutes, and the right-hand bar is for products formed at 250 °C for 60 minutes.

4.3.4 Product use suitability

4.3.4.1 Fuel

For the use of biomass fuels in pulverised fuel boilers, properties similar to coal are desired, particularly the char reactivity and volatile matter content [70]. However, the high alkali and alkali earth metals concentration in the ash of biomass materials have been shown to cause

issues in the boiler [71]. HTC as a biomass treatment can reduce the alkali metal content in the hydrochar compared to the raw biomass, increasing the safe combustion temperature and reducing the theoretical risk of slagging, fouling and corrosion [74].

The ash content of hydrochar is an important parameter for its use as a fuel. Figure 4.10 shows the ash content of the digestate feedstocks and their hydrochars at each of the HTC temperatures. The increase in ash content with increasing HTC severity depends on the particular ash chemistry of the digestate feedstock. It has been concluded that for organic materials, a greater proportion of organic material is removed in the hydrochar than inorganic material, increasing the overall concentration of ash in the product [73]. In order to avoid slagging in the boiler, a high ash melting point temperature is desired. Generally, the presence of potassium and sodium reduce the ash melting temperature, while magnesium and calcium increase it [74]. Figure 4.8 shows that potassium contained in the original feedstock was removed from the hydrochar and mostly recovered in the process liquid fraction. This suggests an increase in ash melting temperature of the hydrochar samples, thus reducing the propensity of the hydrochar to slagging. Fouling is brought about when potassium and sodium combine with chlorine to form alkali chlorides, which condense on heat exchanger surfaces causing corrosion. Therefore, a reduction in potassium concentration of the hydrochar could also reduce the risk of such fouling [73].

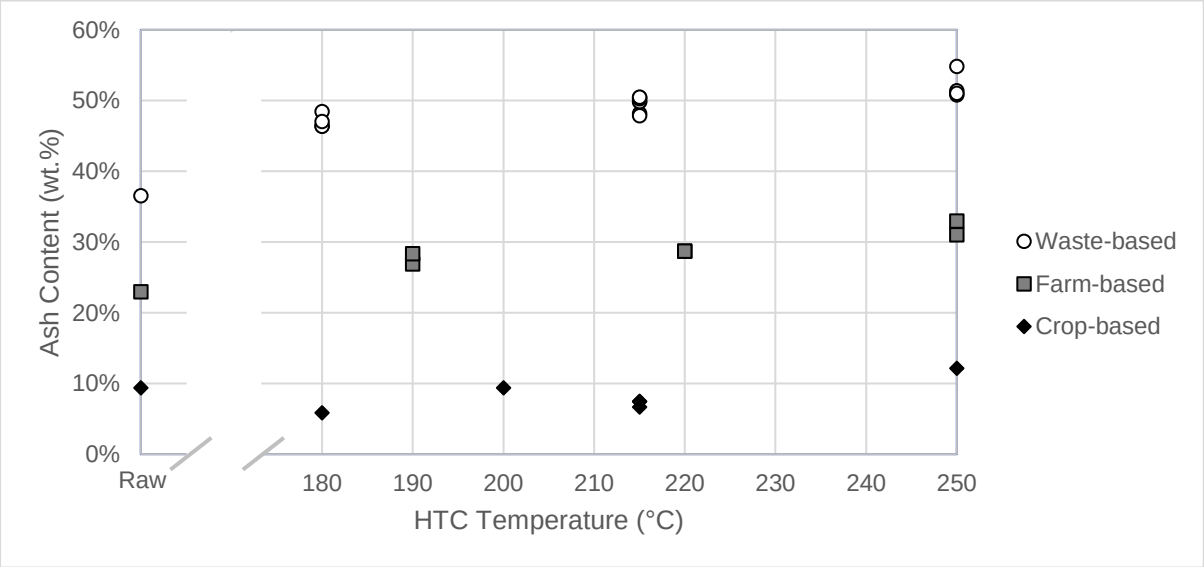


Figure 4.10: Ash content of the raw biomass and hydrochar products for each of the digestate feedstocks.

While the behaviour of the ash in the furnace relates to the specific ash chemistry, high levels of ash are undesirable. On an industrial scale, the removal of large quantities of ash would be a costly and inefficient process. The crop-based digestate is the only feedstock that has a low enough ash content to make it suitable for use as a fuel. Hydrochars higher in ash would

require further post-treatment or would need to be blended with a low ash sample before being considered. For comparison, lignite coal has a high ash content of typically 10–24 wt.%, whereas bituminous coal has a lower ash content of 6–10 wt.% [181]. Lignite is often blended with higher rank coals to improve the overall fuel properties.

Figure 4.11 shows the high heating values (HHV) of each of the digestate and hydrochar samples. This shows how the HHV of the chars increased with an increase in HTC temperature, reaching almost 25 MJ/kg for the crop-based digestate char at 250 °C. This is similar to the heating value of sub-bituminous coal (roughly 24 MJ/kg) and higher than that of lignite (in the region of 15 MJ/kg). Not much of a change in HHV was seen for the waste-based char due to its high ash content. While there was a densification in carbon at higher temperatures (Figure 4.5), the ash content also increased.

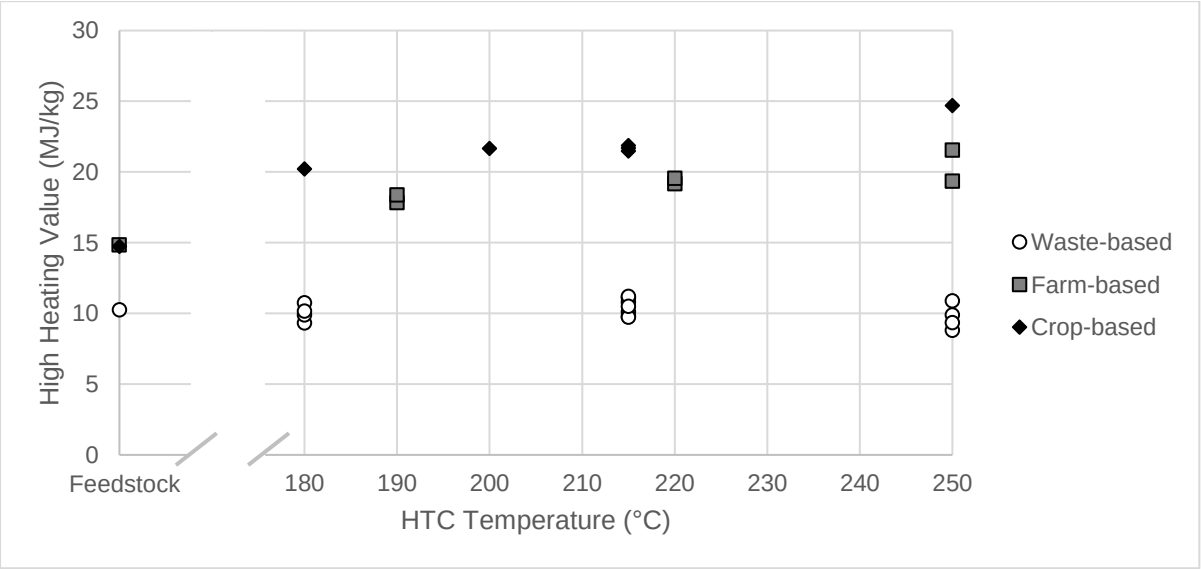


Figure 4.11: High heating value (HHV) on a dry basis of the raw biomass and hydrochar products for each of the digestate feedstocks.

A van Krevelen diagram (Figure 4.12) was plotted as another way of visualising the solid fuel potential of each of the chars. The atomic ratio of H/C and O/C are compared to evaluate the degree of coalification of the chars. The low moisture content of the hydrochars was measured and the associated hydrogen and oxygen contents were subtracted from the results. This illustrates how the properties of the hydrochars are more similar to coal than the raw feedstocks and their likeness increases with an increase in HTC temperature. The crop-based char treated at 250 °C had similar atomic ratios to that of lignite, confirming that this is the most promising solid fuel product, as discussed earlier. In order to fully understand the suitability of hydrochar products for solid fuel, further experimental analysis and investigation into pre/post-treatments is recommended.

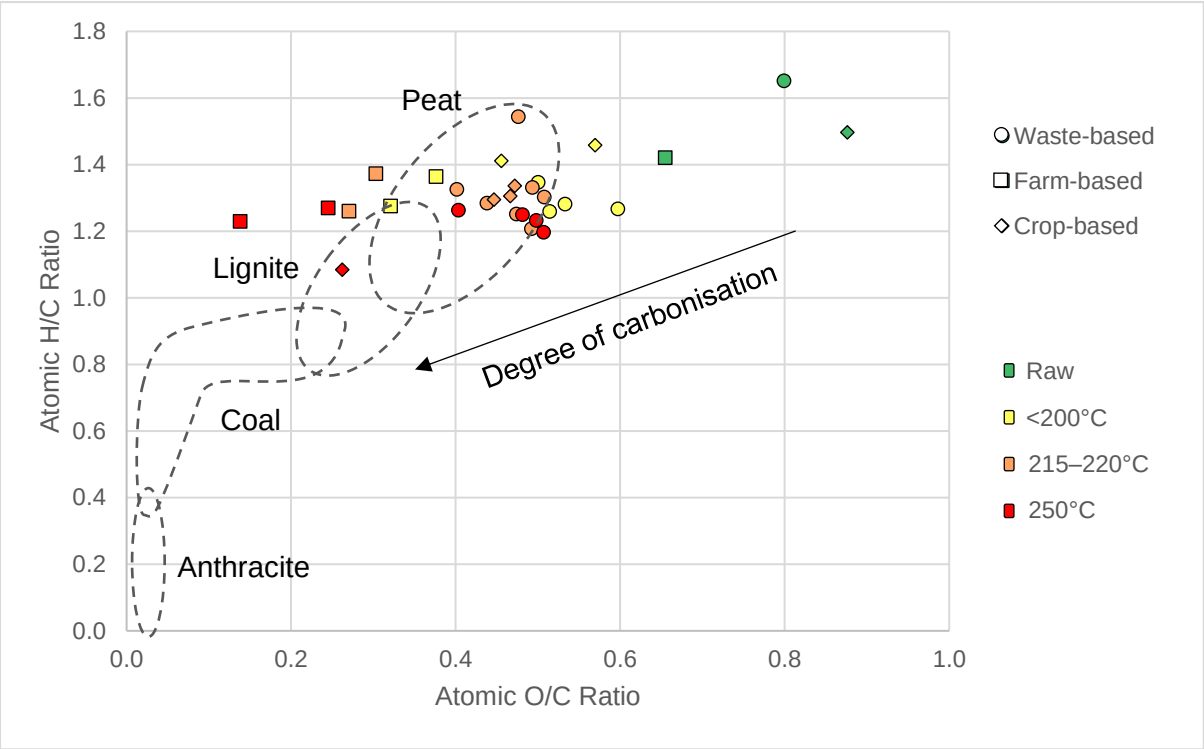


Figure 4.12: Atomic H/C and O/C ratios of the three types of digestate and their derived-hydrochars (Van-Krevelen Diagram). Changes in colour represent the HTC temperature.

4.3.4.2 Fertiliser potential

Digestate, particularly from waste-based and farm-based feedstocks, has a high mineral content that is recovered in the hydrochar and process liquid during HTC. Therefore, interest has been shown in using HTC products as NPK fertilisers [97]. Industrially, process liquid, after desalination and concentration, can be used for the fertigation (the injection of fertilisers into an irrigation system) of agricultural fields [182]. The applicability of this depends on local contracts and regulations, but it remains a beneficial and relatively low-cost business model. Most of the nitrogen and potassium from the digestate is recovered in the process liquid while most of the phosphorus is retained in the solid hydrochar, as seen in Figure 4.8 and Figure 4.9. These results are in general agreement with previous work investigating the fate of plant available nutrients during the HTC of digestate [180]. Methods exist to access the phosphorus retained in hydrochar which could be cost effective as phosphorus is a highly valuable element. However, chemical leaching for phosphorus removal from the hydrochar, for example, has been proven to have an overall negative environmental effect [183].

When assessing the suitability of a product as an NPK fertiliser, the ammoniacal nitrogen is of interest as it is in a plant-available form. The CHN analysis conducted on the digestate feedstocks, hydrochar and process liquid gives the total nitrogen concentration rather than the ammoniacal nitrogen. The waste-based digestate feedstock was sourced from an AD operator

producing digestate to comply with PAS110 standards [164]. In order to meet PAS 110 standards, the AD operator regularly submitted digestate samples for accredited, independent composition analyses to be performed. Results from these independent analyses of the waste-based digestate, including ammoniacal nitrogen content, were made available.

Figure 4.13 shows the nitrogen, phosphorus and potassium concentrations on a ppm basis of the digestates and their HTC products. These results are for HTC runs with process conditions of 215 °C, 180 minutes and an 80 wt.% moisture content for the waste-based digestate, and 220 °C, 180 minutes and an 85 wt.% moisture content for the farm-based digestate, as used in Figure 4.8. 80% of the total nitrogen contained in the waste-based digestate, according to independent PAS110 tests, was in the ammoniacal form. The nitrogen content of the digestate measured using CHN analysis was much lower. The PAS110 tests were performed on untreated, wet whole digestate. However, before the laboratory CHN analysis, the digestate and hydrochar samples were dried for 24 hours. This was necessary to ensure consistency in HTC experimentation. Literature suggests that hydrochar, and presumably digestate, is able to absorb ammonium well, but this sorbed ammonia may then be lost during washing and drying at 105 °C [180]. This would explain the lower reading of N in the digestate measured in the laboratory compared to the independent PAS110 tests. Therefore, it is likely that the raw hydrochar contained more nitrogen than recorded. This study also concluded that the majority of the ammoniacal nitrogen remained on the hydrochar. Therefore, while the ammoniacal nitrogen was not measured in the laboratory, the hydrochar products could contain valuable quantities of plant-available nitrogen. A sensitivity analysis is used when considering the economic potential of HTC products in the fertiliser market (Chapter 6 Case Study).

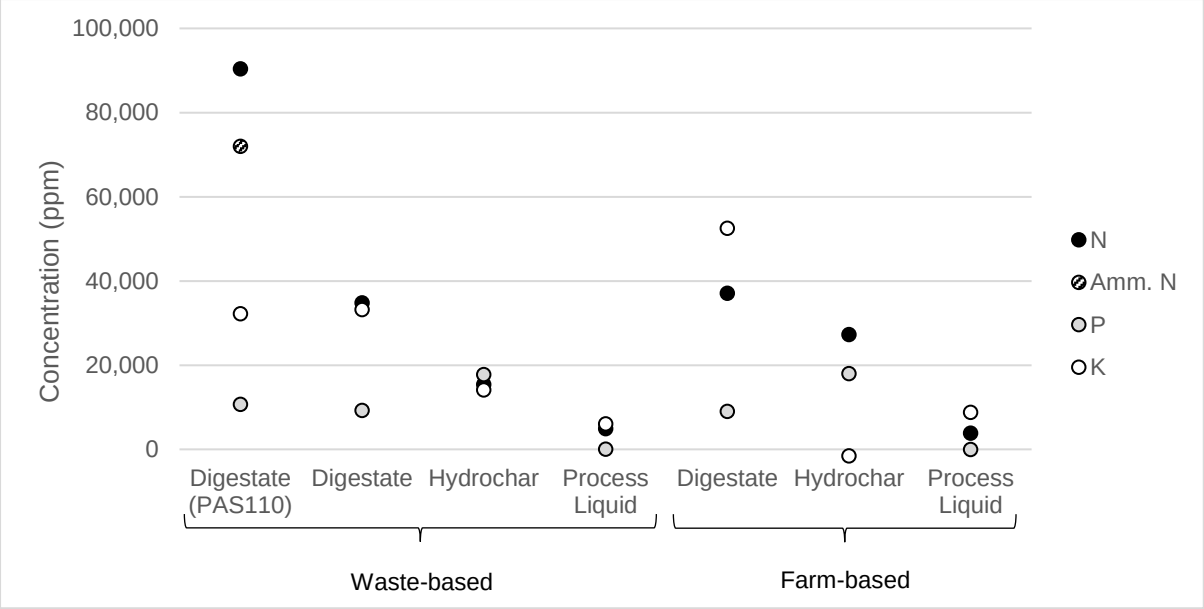


Figure 4.13: Concentration of fertiliser nutrients in the digestate, hydrochar and process liquid samples for the waste-based and farm-based digestates.

4.3.4.3 Soil application

When considering hydrochar in agricultural use, the amount of carbon that can be stored is of high interest. Soil application has been proposed as a method of carbon sequestration and the concentrations shown in Figure 4.5 give an indication of how much carbon could be stored in land. There is limited data on the long-term effects of the application of hydrochar to land, with mixed results. One study concluded that raw hydrochar degrades over tens of years making it a suitable candidate for carbon sequestration [95]. Another found that hydrochar was very labile, due to its low preparation temperature compared to charcoal, and so it was unsuitable for long-term sequestration. They did not rule out the application of hydrochar to land, however, and stated a potential use as a slow-release fertiliser product [96]. Further long-term field experiments of both raw and treated hydrochars are recommended to better understand its suitability. The heavy metals concentration in hydrochar must also be considered. There are existing regulations for biochar materials and so the waste-based and farm-based hydrochar products have been compared to these. The European Biochar Certificate (EBC) was created in 2013 to standardise the use of biochar in agriculture [92]. The Biochar Quality Mandate (BQM) is a similar set of standards developed in the UK [170]. The results for the waste-based and farm-based digestates and resulting hydrochars are shown in Table 4.3. Again, the hydrochars produced at 215 °C, 180 minutes and an 80 wt.% moisture content for the waste-based digestate, and 220 °C, 180 minutes and an 85 wt.% moisture content for the farm-based digestate were used.

Table 4.3: Heavy metal content (ppm, mg/kg) compared to the standards detailed in the EBC and BQM. Values in brackets are for premium/high-grade quality biochar. Green represents full compliance, yellow represents compliance with the basic/low quality limits and orange represents compliance with only the EBC and/or BQM and not both. Where no results are available, grey is used. Below LoD indicates that the concentration was below the limits of detection.

		As	Cd	Cr	Cu	Hg	Mn	Mo	Ni	Pb	Se	Zn
	EBC	13	1.5 (1)	90 (80)	100	1			50 (30)	150 (120)		400
	BQM	100 (10)	39 (3)	100 (15)	1500 (40)	17 (1)	3500	75 (10)	600 (10)	500 (60)	100 (5)	2800 (150)
Waste-based	Dig.		1	31	32		69	5	18	9	Below LoD	993
	Char		1	58	63		133	9	32	16	Below LoD	1916
Farm-based	Dig.		1	11	36		193	4	16	3	Below LoD	251
	Char		1	23	76		400	8	34	6	Below LoD	524

Generally, both the digestates and hydrochars fell within the concentration limits set out in the standards, with only Zn exceeding the EBC regulations. Raw hydrochar has been shown to contain phytotoxic compounds and so it is expected that it would undergo further treatment before land application. One study demonstrates how a further thermal treatment at 275 °C gives the best trade-off in terms of phytotoxic compound removal and mass yield [86]. This treatment led to noticeable improvements in crop growth but resulted in a 25% mass loss and a relative increase in ash content. The results in Table 4.3 do not account for this post-treatment. If the hydrochar had undergone this treatment, the heavy metal contents would increase on a ppm basis due to the relative increase in ash. However, as the majority of elements fall well below the limits, this is not envisaged as a hurdle to agricultural use.

4.3.4.4 Plant trials

Independent plant growth trials were carried out at the Biorenewables Development Centre, based at the University of York. Due to the large quantity of char required in the plant trials, 5 litres of hydrochar was sourced from CPL Industries. The hydrochar had been produced from food waste digestate provided by GWE Biogas, an AD operator processing 50,000 tonnes/year of commercial food waste. This digestate was assumed to be most similar to the waste-based digestate sample due to its food waste origin.

Previous studies that hydrochar is phytotoxic in nature, with raw hydrochar resulting in germination inhibition [80,81]. Various hydrochar post-treatments have been shown to reduce

the phytotoxicity of hydrochar, be that physical [80,89], biological [81,84,85], chemical [86–88], or thermal [86,87]. More details of the suitability of hydrochar can be found in Section 2.4.2 Hydrochar soil application.

Despite the foreseen hurdles and unknowns, the annual generation of digestate in the UK contains somewhere between 133,000 and 240,000 tonnes of carbon (Table 6.1, Section 6.2 AD in the UK) and therefore there is interest in the potential to store this carbon in land, both from an environmental and a financial viewpoint. These plant trials were performed to investigate if the application of thermally treated hydrochar from digestate resulted in complete inhibition of crop germination or whether it was permissible at certain concentrations, offering up land application for carbon storage as a potential market.

Raw hydrochar has been shown to contain phytotoxic compounds and so a thermal treatment was used prior to plant trials. The hydrochar was heated to 275 °C at a rate of 3 °C/min and held for 4 hours under vacuum, based on the optimum conditions by Hitzl et al. [86]. Following this, the treated hydrochar was mixed with coir in varying concentrations on a volume basis and basil seeds were sown as part of 2-week germination trials which extended to 8-week plant growth trials. Three trays were used for each hydrochar concentration.

Figure 4.14 shows the results for one tray from each of the concentrations and is representative of the three trays used. This shows that while higher hydrochar concentrations led to a decline in plant health and growth, no severe inhibition was observed until the concentration reached 50 vol.%. Figure 4.15 shows the average number of true leaves for each of the samples. Generally, there was a steady increase in the number of leaves over the time period. This shows that the 0 vol.% hydrochar (and thus 100 vol.% coir) sample resulted in plants with the highest number of leaves. By the end of the 8 weeks, there was only a small difference in the number of leaves between the 0 vol.% and 10 vol.% hydrochar samples. However, it is clear from Figure 4.14 that the plants with low concentrations (10 vol.% and 20 vol.%) have poorer general health and vigour. Figure 4.16 shows the average plant height and total dry weight at the end of 8 weeks. Again, this confirms that the healthiest sample was the sample containing 0 vol.% hydrochar, but there was little difference in the samples containing 10 and 20 vol.%.

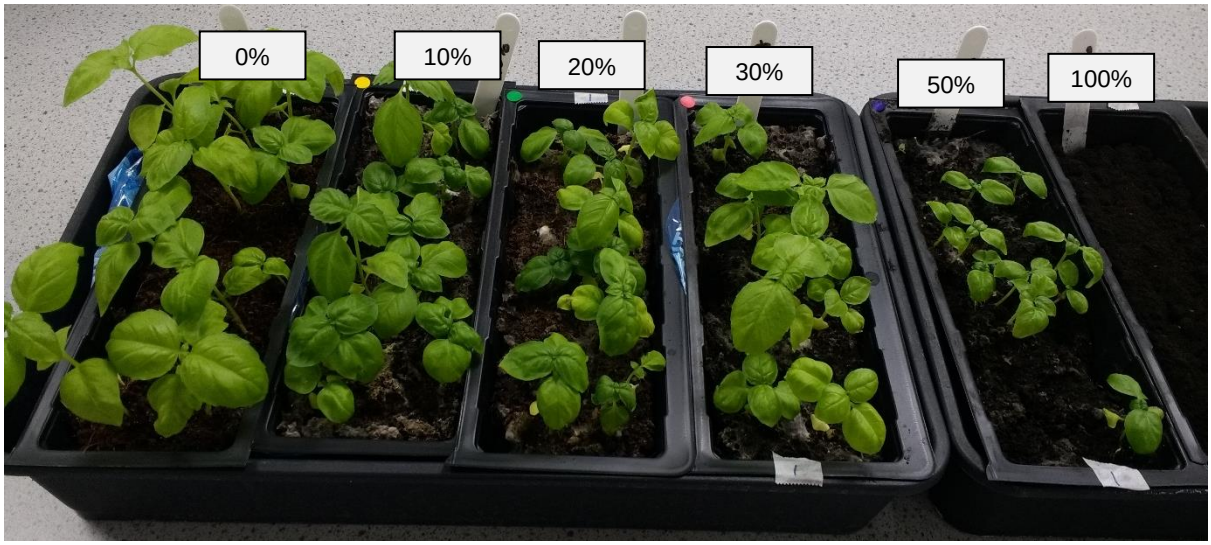


Figure 4.14: Tray 1 for each of the concentrations of hydrochar (vol.%) after 8 weeks.

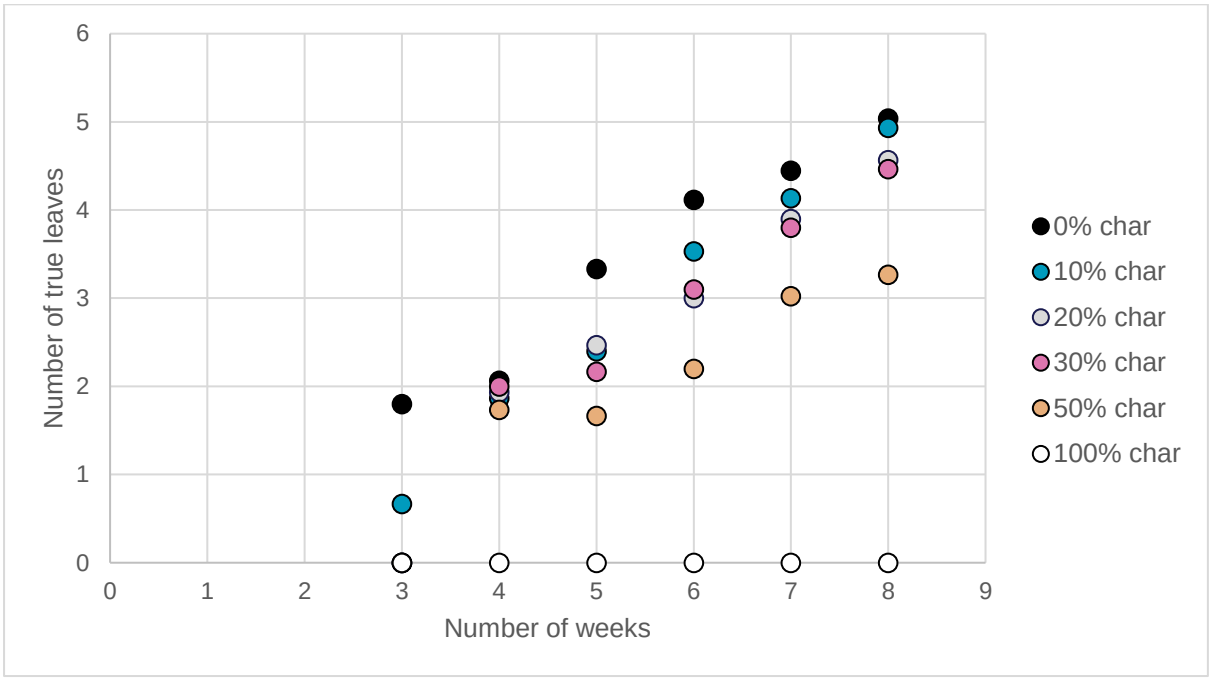


Figure 4.15: Average number of true leaves per plant for each concentration of hydrochar (vol. %).

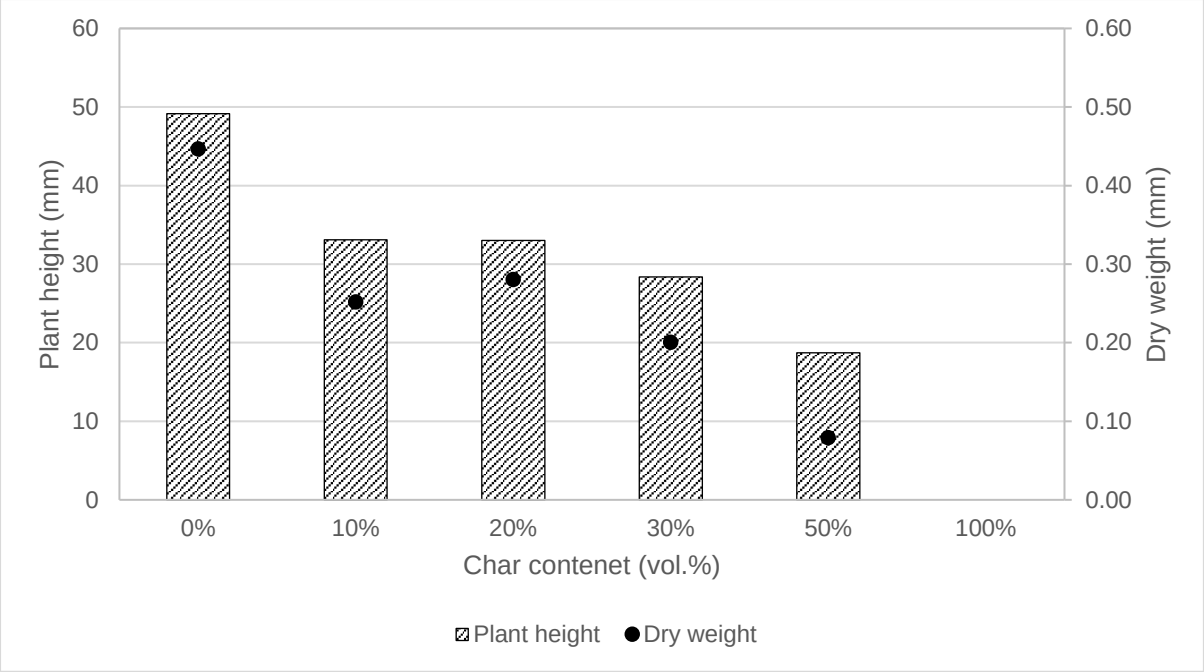


Figure 4.16: Average plant height and overall dry weight of each of the samples at the end of the 8-week period.

The results of the plant growth trials suggest that the inclusion of the treated hydrochar in the growth media did not result in complete inhibition. An independent fertiliser analysis was carried out for the treated hydrochar and there appeared to be much lower levels of plant nutrients than for the waste-based hydrochar used in the laboratory. Phosphorus was the only element above the limits of detection in the fertiliser analysis, with its concentration converting to 3000 ppm of dry char, 5 times lower than the 15,000 ppm measured in the waste-based hydrochar. A reduction in nutrient content is likely to have occurred in the thermal post-treatment of the hydrochar and further analysis is recommended to better understand this.

4.4 Regression model

4.4.1 Development of model

Regression analysis was used for each of the types of digestate to predict HTC product properties from the process conditions studied. For the waste-based digestate, a full Box-Behnken design of experiments was performed. The Response Surface Design on Minitab was used to analyse the results. The backwards elimination method was used to remove any variables that had a significance (p-value) of less than 0.05. For the farm-based and crop-based digestates, a complete design of experiments was not performed and so a regression model was fitted using Minitab using the backward elimination method with a minimum significance of 0.05.

The Response Surface Design and regression models were able to predict the mass yields of the different product fractions and their composition based on the HTC processing conditions. For the composition, both the elemental recovery and weight fraction were calculated. The equation which gave the highest R^2 predictive value was used by the model to predict the composition of the products. For example, the regression model for the waste-based digestate gave a reasonable fit for the carbon recovery (R^2 of 82.9% and R^2 predictive of 65.9%) but the hydrochar carbon content did not result in any significant terms. Therefore, the carbon recovery regression was added to the model and the resulting hydrochar carbon content was calculated based on that. Table 4.4 lists the response variables that were used in the regression model.

Table 4.4: Response variables modelled by the regression models. The P and K hydrochar contents were only modelled for the waste-based and farm-based digestate materials.

Type	Mass yield	Hydrochar elemental composition	Hydrochar proximate composition
Products	Solid, liquid and gas	C, H, N, O, (P and K)	VM, FC and Ash
Variants	Dry and dry ash-free	Dry, dry ash-free and recovery	Dry and recovery

4.4.2 Model results

The statistical model supported previous observations of experimental and literature results. Table 4.5 gives a general overview of the significance and size of the effect of each of the factors studied. In every regression model, temperature was shown to have the largest effect. This is in agreement with previous work on HTC severity [137]. Time was only shown to have a significant effect in a small number of models. Moisture content, however, was shown to be significant for the majority of models for the waste-based digestate. Interaction effects were not included in the simple regression models of the farm-based and crop-based digestates. However, interaction effects were shown to be significant for a number of models for the waste-based digestate. A comprehensive comparison of the model developed here with literature models is included in Section 4.4.4. The effect of process conditions on the economics of the HTC process are explored in Chapter 5 Process Model. An increase in moisture content, in particular, led to a large increase in plant size and thus a significant increase in capital cost. This is something that must be considered by plant operators when selecting optimal HTC plant conditions.

Table 4.5: An overview of the significance and size of effect for each of the factors.

	Temperature	Time	Moisture content	Interaction effects
Waste-based	Always significant, always has the largest effect	Only for P recovery	Significant in most cases	Often significant, notably T^2, M^2 and T*M
Farm-based		Significant for some elements of hydrochar	N/A	N/A
Crop-based		Never significant	Never significant	N/A

A regression model was generated for each of the terms (solid yield, carbon content, ash content etc.) on percentage recovery, raw, dry-ash free and/or dry bases. Of these, the regression equation which gave the highest R^2 predictive value was selected as the best estimate and subsequently used in the process model. The regression models used are shown in Table 4.6. The use of regression models in the prediction of HTC products and characteristics is explained in Chapter 2 Literature Review Section 2.3.2. The general form of a regression equation is shown in Eq. 2.1.

$$Y_i = \alpha + \beta_1 X_{i,1} + \cdots \beta_p X_{i,p} \qquad \textit{Eq. 2.1}$$

where α is a constant, β is the coefficient and X is the factor of which there are i number.

As an example, to estimate the solid mass fraction of products (i.e., hydrochar produced) on a dry, ash-free basis for the waste-based digestate, the following equation was used:

$$Y_{solid,daf} = -1.41 - 0.0035\mathrm{T} + 5.49\mathrm{M} - 0.0038\mathrm{TM} + 1.35\times10^{-5}\mathrm{T}^2 - 2.767\mathrm{M}^2 \text{ (Table 4.6)}$$

It is not possible to compare the effect of each factor based on their coefficients from this equation alone. The magnitudes of the coefficients are relative to the units of the factor itself and interactive effects also impact the final result also. The relative importance of each factor and interaction effect is investigated in Chapter 5 HTC Plant Model Section 5.6 Optimisation.

The waste-based digestate tended to give better estimates (R^2 adjusted value) for product yield fractions. A full Box-Behnken design of experiments was performed for this type of digestate whereas only 6 sets of design conditions were used for the farm-based and crop-based digestates. This indicates that a higher number of experimental runs results in estimates with greater accuracy, as expected in Section 4.2 Feedstock. However, this trend is not observed for the C value of the waste-based digestate. A possible explanation for this is that the waste-based hydrochar has a lower carbon content than the farm-based and crop-based digestate. Therefore, inaccuracies in the experimental measurement of carbon will have

represented a higher percentage error in the final reading. This theory is supported by the inability to generate a regression model containing significant terms for the low-ash crop-based sample, whereas reasonable models for ash were found for the higher-ash waste-based and farm-based hydrochars. Overall, the estimate and use of these regression models were deemed acceptable for the purpose of this general study. If better estimates were desired, either a larger number of data points or more sensitive analysis methods are recommended.

Table 4.6: The adjusted R^2 value (percentage variation in the response), S (standard error of the regression) and the regression coefficients for the three types of digestate. T is HTC temperature (°C), t is residence time (minutes), and M is moisture content (m.f.). m.f. is mass fraction, e.g. a mass fraction of 0.5 represents a 50 wt.%. Only significant values (p-value<0.05) are included, insignificant terms are left blank. Where the experimental runs did not explore a particular factor or interaction effect, N/A is used. As explained in Chapter 4 HTC Test Programme, each term was calculated a number of ways (dry, daf, recovery etc.) and the equation which gave the highest R^2 predictive value was used, detailed here.

	Term	Units	R^2 (adj.)	S	Constant	T	t	M	T*t	T*M	t*M	T^2	t^2	M^2
Waste-based	Solid	m.f. (daf)	94.1%	0.009	-1.41	-0.0035		5.4900		-0.0038		1.30E-05		-2.7670
	Liquid	m.f. (daf)	94.7%	0.008	2.81	0.0020		-6.0900		0.0060		-1.50E-05		2.9100
	Gas	m.f. (raw)	85.1%	0.007	-0.66	0.0039		0.6710		-0.0040				
	VM	% recovery	97.6%	0.005	-1.15	-0.0038		5.0150		0.0031		1.30E-05		-2.5840
	FC	No significant terms found, calculated by difference using VM and ash.												
	Ash	m.f. (dry)	76.3%	0.011	0.24	0.0007		0.1207						
	C	% recovery	78.2%	0.016	-2.78	-0.0010		8.3000						-4.8800
	H	% recovery	54.3%	0.028	-4.26	-0.0010		11.3300						-6.5500
	N	% recovery	84.2%	0.020	-2.40	-0.0015		7.1500						-4.2800
	O	% recovery	72.2%	0.021	1.67	-0.0115						2.40E-05		
	P	% recovery	77.1%	0.004	0.98	0.0002	0.0001	-0.0546						
	K	ppm (liq.)	89.8%	620	28381	15		-32016						
Farm-based	Solid	m.f. (daf)	68.9%	0.035	0.96	-0.0020		N/A	N/A	N/A	N/A	N/A	N/A	N/A
	Liquid	No significant terms found, calculated by difference using solid and gas yield.												
	Gas	m.f. (daf)	99.0%	0.003	-0.23	0.0013	0.0001	N/A	N/A	N/A	N/A	N/A	N/A	N/A
	VM	m.f. (daf)	95.0%	0.008	1.01	-0.0010	-0.0003	N/A	N/A	N/A	N/A	N/A	N/A	N/A
	FC	m.f. (daf)	96.0%	0.007	-0.01	0.0010	0.0003	N/A	N/A	N/A	N/A	N/A	N/A	N/A
	Ash	m.f. (dry)	75.9%	0.011	0.13	0.0007		N/A	N/A	N/A	N/A	N/A	N/A	N/A
	C	wt.% (dry)	88.4%	0.008	0.28	0.0007	0.0002	N/A	N/A	N/A	N/A	N/A	N/A	N/A
	H	% recovery	92.1%	0.014	0.97	-0.0015	-0.0004	N/A	N/A	N/A	N/A	N/A	N/A	N/A
	N	No significant terms found, calculated by difference using C, H and N concentrations.												
	O	% recovery	93.0%	0.024	1.01	-0.0030	-0.0006	N/A	N/A	N/A	N/A	N/A	N/A	N/A
	P	% recovery	61.0%	0.007	0.91	0.0004		N/A	N/A	N/A	N/A	N/A	N/A	N/A
	K	No significant terms found, calculated by linear interpolation												
Crop-based	Solid	m.f. (raw)	82.0%	0.041	1.42	-0.0039			N/A	N/A	N/A	N/A	N/A	N/A
	VM	m.f. (daf)	78.7%	0.038	1.40	-0.0032			N/A	N/A	N/A	N/A	N/A	N/A
	FC	m.f. (daf)	85.3%	0.023	-0.32	0.0024			N/A	N/A	N/A	N/A	N/A	N/A
	Ash	No significant terms found, calculated by difference using VM and FC.												
	C	wt.% (dry)	92.3%	0.010	0.22	0.0015			N/A	N/A	N/A	N/A	N/A	N/A
	H	wt.% (dry)	80.7%	0.001	0.08	-0.0001			N/A	N/A	N/A	N/A	N/A	N/A
	N	wt.% (dry)	74.4%	0.001	-0.01	0.0001			N/A	N/A	N/A	N/A	N/A	N/A
	O	wt.% (dry)	80.6%	0.024	0.78	-0.0022			N/A	N/A	N/A	N/A	N/A	N/A

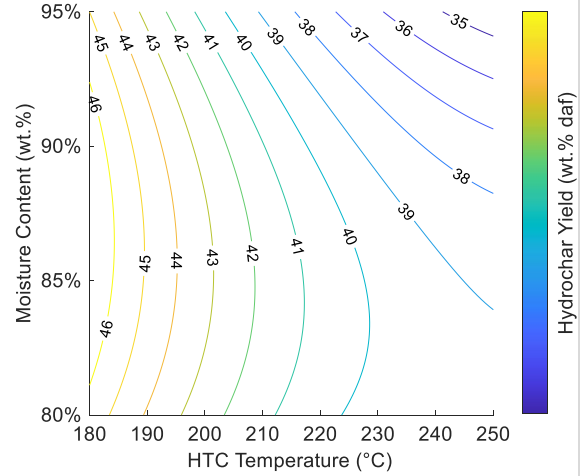
Figures 4.17, 4.18 and 4.19 show the model results for hydrochar yield (daf), the hydrochar carbon concentration (daf) and the ash content of the hydrochar for all three of the digestate feedstocks used. The waste-based digestate and crop-based digestate were plotted for the HTC temperature and the feedstock moisture content. The farm-based digestate was plotted for the HTC temperature and residence time. The axes reflect the factors that were found to be significant in the model, using a p-value of <0.05 as a basis. Residence time was not significant in any of these models for the waste-based and crop-based digestates. Moisture content was held constant and therefore was not a factor for the farm-based digestate. R^2 is the percentage of variation in the response that is explained by the model. The adjusted R^2 value incorporates the number of predictors in the model to allow for comparison between the feedstocks. S is the standard error of the regression, the average distance of the data points from the fitted model. A high R^2 value and low S indicate a model with a good fit.

For the hydrochar yield (daf) (Figure 4.17), each of the models show a decrease in yield with an increase in HTC reaction temperature. For the waste-based digestate, the moisture content and interaction effects were also shown to affect the yield to a lesser degree. This trend was seen and discussed earlier in Figure 4.4. The surface plots show a generally good agreement with the experimental data points. A linear trend, only depending on HTC temperature is shown for the farm-based and crop-based digestates.

The carbon concentration (daf) of the hydrochar (Figure 4.18) increased with an increase in process temperature. Again, this trend was seen and discussed earlier in Figure 4.5. The hydrochar from the waste-based digestate showed a significant relationship with moisture content, and a larger effect still was seen with the moisture content squared. This explains the curving of the contour plot. Residence time was found to significantly affect the hydrochar composition for the farm-based digestate, but it clearly had less of an effect than the HTC temperature. The crop-based hydrochar was only affected by the HTC temperature. The regression models gave good estimates of hydrochar carbon composition, as reflected by the high values of R^2 (adj.) and low values of S.

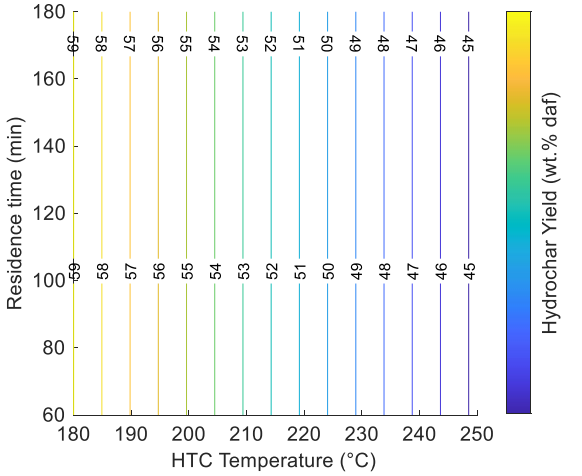
The ash content of the hydrochar, again, was largely affected by HTC temperature for each of the feedstocks (Figure 4.19). Only the waste-based hydrochar appeared to be affected by an additional factor, namely moisture content. While HTC temperature was shown to have a significant effect on hydrochar ash content, it is clear that the original feedstock had a much greater influence than the HTC process conditions. The fit of the regression models for ash content was not as good as the hydrochar yield and carbon content, reflected in the lower R^2 and higher S values. The ash content had to be calculated by difference for the crop-based digestate.

a) Waste-based digestate:



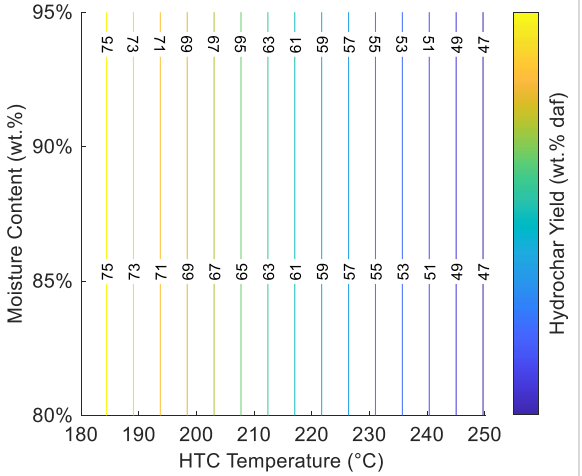
R^2 (adj.) = 94%, S = 0.009

b) Farm-based digestate



R^2 (adj.) = 69%, S = 0.035

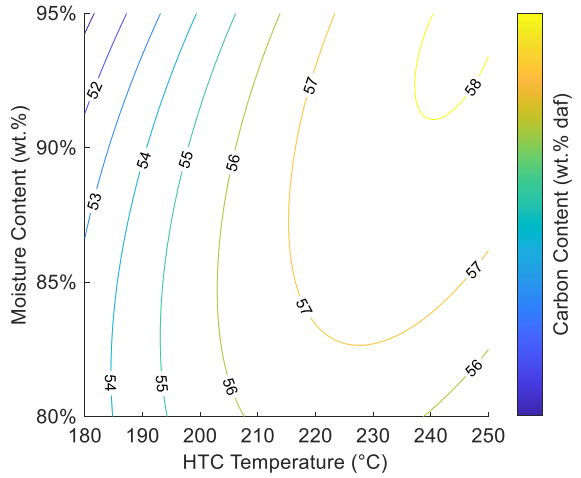
c) Crop-based digestate



R^2 (adj.) = 83%, S = 0.043

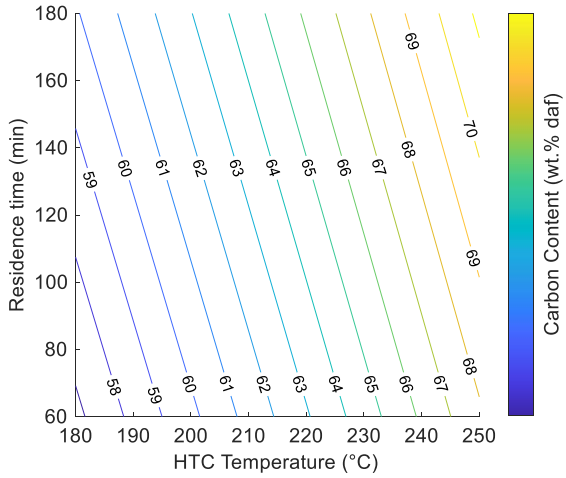
Figure 4.17: Hydrochar yield on a dry ash-free basis for each of the digestate feedstocks. Note that the waste-based and crop-based digestates are plotted against moisture content, but the farm-based digestate is against residence time.

a) Waste-based digestate:



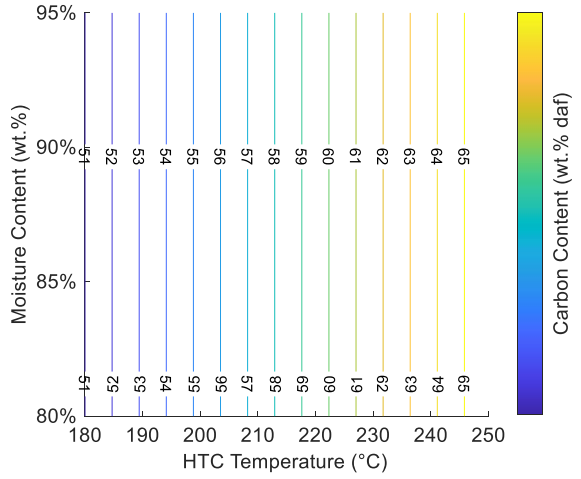
R^2 *(adj.) = 78%, S = 0.016*

b) Farm-based digestate



R^2 *(adj.) = 88%, S = 0.008*

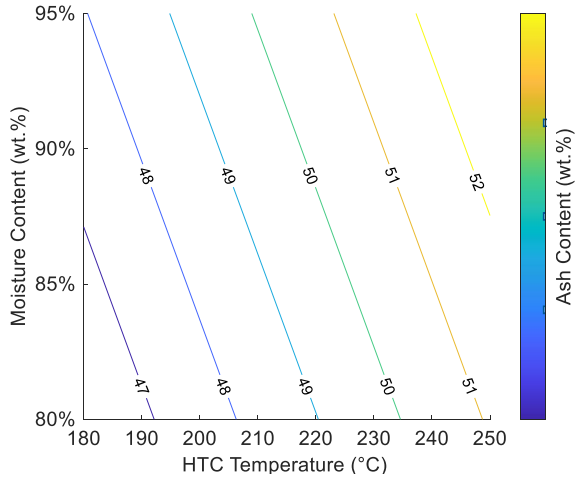
c) Crop-based digestate



R^2 *(adj.) = 92%, S = 0.010*

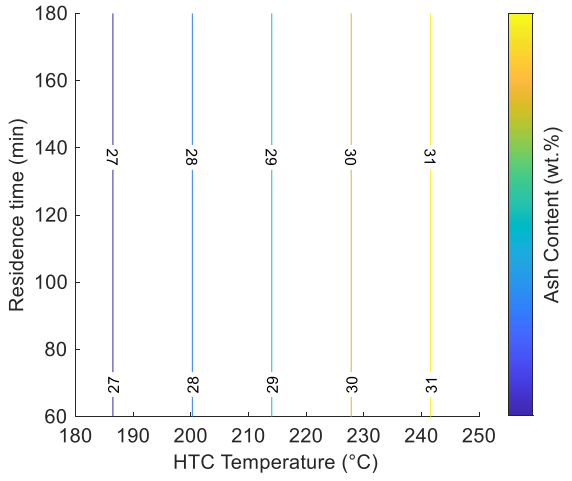
Figure 4.18: Hydrochar carbon content on a dry ash-free basis for each of the digestate feedstocks. Note that the waste-based and crop-based digestates are plotted against moisture content but the farm-based digestate against residence time.

a) Waste-based digestate:



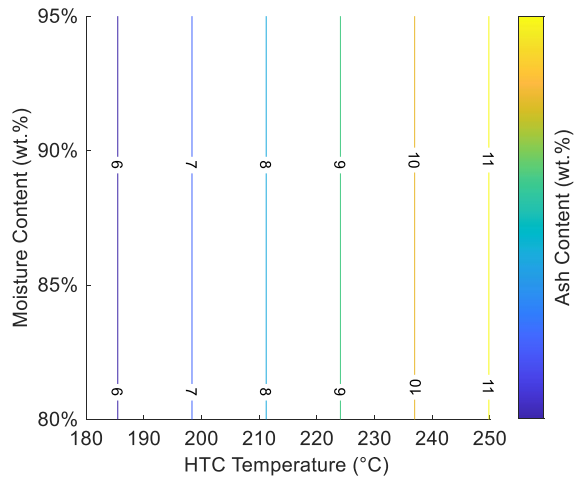
R^2 (adj.) = 76%, S = 0.011

b) Farm-based digestate



R^2 (adj.) = 76%, S = 0.011

c) Crop-based digestate



calculated by difference using the VM and FC content with R^2 (adj.) = 79%, S = 0.038 and R^2 (adj.) = 85%, S = 0.023 respectively

Figure 4.19: Hydrochar ash content, dry basis, for each of the digestate feedstocks. Note that the waste-based and crop-based digestates are plotted against moisture content but the farm-based digestate against residence time.

4.4.3 Repeatability

The Box-Behnken design of experiments for the waste-based digestate used 3 centre-point runs at the same conditions. In order to explore the repeatability of the HTC experimental procedure, a further 2 runs were carried under the same conditions. It was expected that these 5 runs would give a good representation of the repeatability for all conditions and feedstocks used.

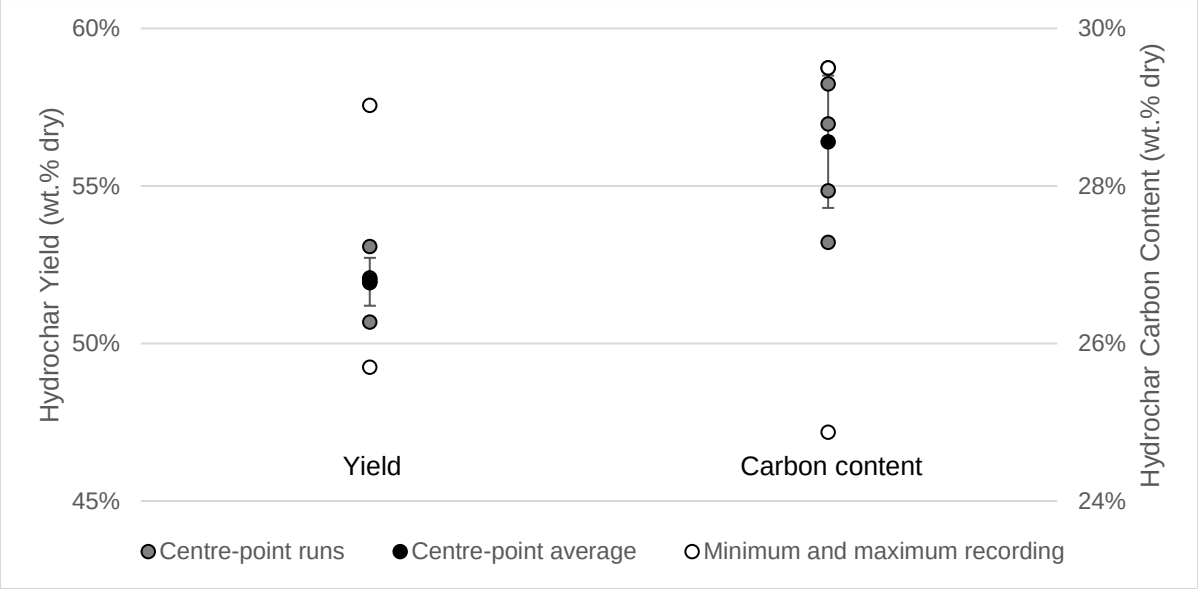


Figure 4.20: The mean value and standard deviation for the centre-point runs repeated 5 times of the solid hydrochar yield and hydrochar carbon content on a dry basis. The maximum and minimum results of the results taken at all process conditions are indicated to show the range of values. Note that the y axis does not extend to zero in order to display the results clearly.

Figure 4.20 shows the mean value and standard deviation of the centre-point runs for hydrochar yield and hydrochar carbon content on a dry basis. Due to laboratory access restrictions, the hydrochar ash contents of the further 2 runs carried out to explore repeatability could not be measured and therefore the mean and standard deviation on a dry ash-free basis could not be measured.

The standard deviation for the hydrochar yield was 0.84 wt.% and the standard deviation of the mean was 0.38. For the hydrochar carbon content, the standard deviation was 0.84 wt.% and the standard deviation of the mean was 0.37. It can be reasonably assumed that the standard deviation for hydrochar yield and hydrochar carbon content would be similar at all other process conditions.

The predictive capability of the regression model was tested by using the 2 repeat readings that were not used in its generation. Figure 4.21 compares experimentally measured and predicted values (generated using the regression models) for hydrochar solid yield and hydrochar carbon content on a dry basis. The solid line represents a 1:1 relationship between the two. In theory, the results of a perfect regression model with a R^2 of 1 would fit to this line. The 2 repeat readings are coloured in grey and it can be seen that they are relatively close to the solid line. This indicates that the regression models (Table 4.6) have a good predictive capability. The percentage error of the experimental value compared to the predicted value was on average 1.3% for the hydrochar yield and 1.1% for the hydrochar carbon content.

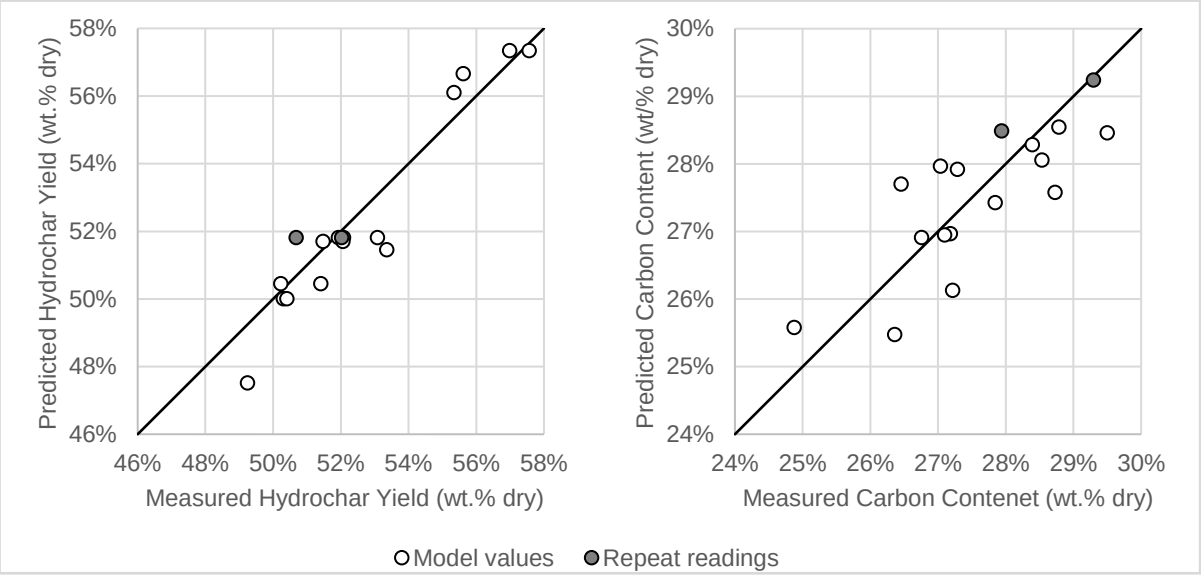


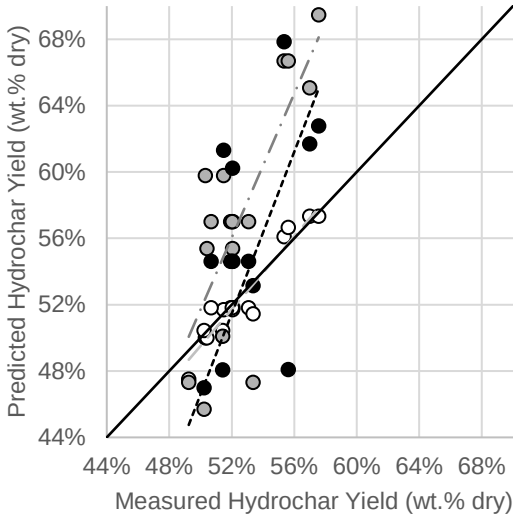
Figure 4.21: Comparison of experimental and predicted values of hydrochar yield and hydrochar carbon content of a dry basis for the waste-based digestate. The solid line represents a 1:1 relationship. The 2 repeat readings taken at the centre-point process conditions, not used in the derivation of the regression model, are coloured in grey.

4.4.4 Comparison with literature models

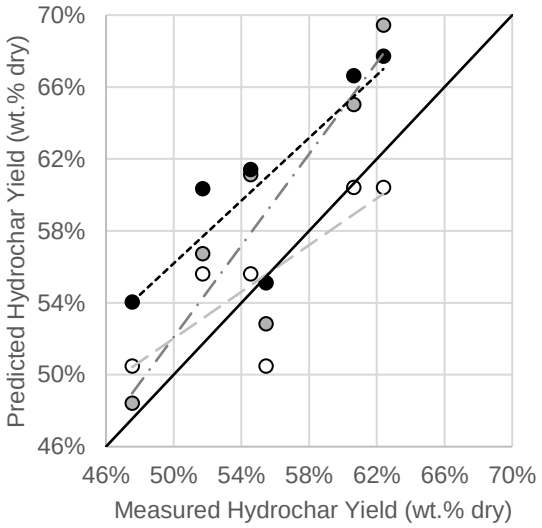
Another method of assessing the goodness of fit of the model is comparing it to meta-analysis data found in literature. The statistical tool developed by Li et al. [65] aimed to create a fully flexible model with which the feedstock properties and process conditions were used to determine the yields and energy contents of the products. They presented regression tree models alongside multiple linear regression equations. Vallejo et al. [67] conducted a similar study, this time accounting for lignocellulosic and non-lignocellulosic feedstocks through the inclusion or omission of the reactivity index, respectively.

The multiple linear regression models from these two literature sources were compared with the model developed in this work. The regression models were used to predict the hydrochar yield (dry basis) for the three types of digestate feedstock. Figure 4.22 compares these predictions to the experimentally measured yields. Neither the Li et al. model [65] nor the Vallejo et al. model [67] gave a good estimate of hydrochar yield for the waste-based digestate used in this work. For the farm-based digestate, the literature models gave a reasonable estimate, which tended to be a slight overestimate. Conversely, the literature models gave a slight underestimate for the crop-based digestate. In both cases, the Vallejo et al. model matched the experimental results better than Li, shown by the proximity of the trendline to the 1:1 ratio. As expected, the regression model developed in this work gave the best results. Li et al. reported that feedstock properties had a contribution of 34% to the calculated mass yield in their model, with process conditions accounting for the rest. Vallejo et al. used the polarity index in their model, a ratio based on the elemental content of the feedstock.

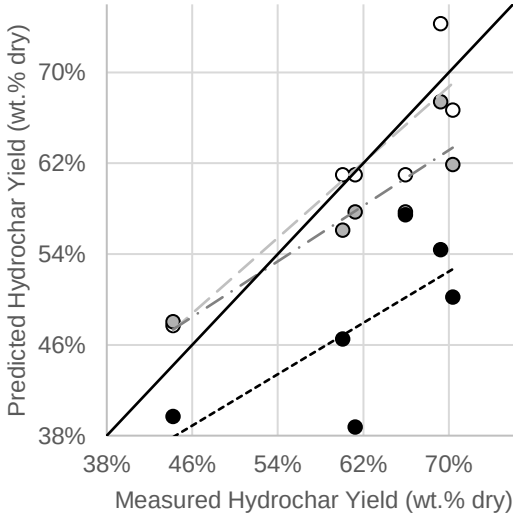
a) Waste-based digestate:



b) Farm-based digestate



c) Crop-based digestate



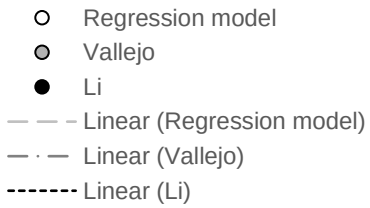


Figure 4.22: A comparison of the regression model developed in this work and predictive models from literature for each type of digestate feedstock [65,67]*. The solid line represents a 1:1 relationship.*

While both models accounted for feedstock properties, it could be that they gave poor estimates for the waste-based digestate due to its significantly larger ash content, of almost 40 wt.%, than most feedstocks typically studied using HTC. For the three digestate feedstocks tested experimentally, a higher ash content resulted in lower hydrochar yields (Figure 4.4). Deriving a regression model for the yield on a dry ash-free basis could result in a more accurate generic model. To test this theory, a simple regression model was generated using Minitab for the yield on a dry and dry ash-free basis for the three digestate feedstocks used experimentally. The process conditions and feedstock ash and carbon content were used as

factors. The same regression method as used for the farm-based and crop-based digestates was followed, using a backward elimination method with a minimum significance of 0.05.

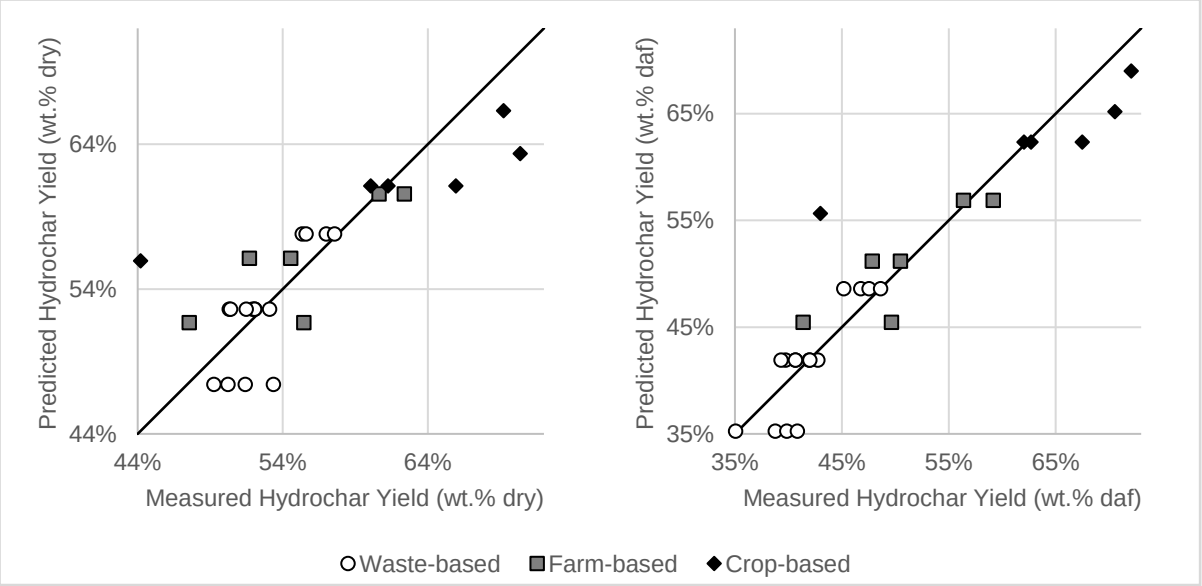


Figure 4.23: Comparison of experimental and predicted values of hydrochar yield on a a) dry basis and b) dry ash-free basis for all three types of digestate feedstocks. The solid line represents a 1:1 relationship.

The models for both the dry and dry ash-free hydrochar mass yield only contained the process temperature and feedstock ash content as significant factors. However, as a consistent design of experiments was not performed for all feedstocks, interaction effects were not considered in this model. Figure 4.23 depicts the goodness of fit of these regression models. It can be seen from these that the relationship was better for hydrochar yield on a dry ash-free basis; this is supported with R^2 values of 66% on a dry basis and 87% on a dry ash-free basis. Therefore, for any development of a generic regression model to be used for different feedstocks, using a dry ash-free basis could give better results for certain property types.

4.5 Conclusions

In accordance with literature findings, HTC temperature had the largest effect on experimental results compared with residence time and moisture content. An increase in HTC temperature led to a decrease in hydrochar yield but an increase in its carbon content (Figure 4.4 and Figure 4.5). The experimental procedure was deemed repeatable, through use of 5 repeat centre-point runs.

The measured recovery of carbon, phosphorus and potassium in the HTC products generally fell between the range of 90%–110% (Figure 4.6, Figure 4.8 and Figure 4.9). However, the recovery of nitrogen ranged from anywhere between 60% and 140% (Figure 4.6). This

discrepancy could be due to the adsorption and subsequent loss of nitrogen during the sample preparation of the feedstock and hydrochar products.

When considering the HTC product markets, the hydrochars of the waste-based and farm-based digestates were deemed unsuitable as solid fuels due to their high ash contents. However, hydrochar from the crop-based digestate at high HTC temperatures showed properties similar to lignite (Figure 4.12). Aside from potassium and sodium, the inorganic elements contained in the digestate feedstocks almost wholly remained in the solid hydrochar post-HTC (Figure 4.8). Hydrochar samples contained respectable quantities of nitrogen and phosphorus, indicating their potential use in the fertiliser market. Potassium levels tended to be lower, but the process liquid could prove as a useful source. Generally, both the digestates and hydrochar complied with the heavy metal concentration limits for soil application set out by international standards (Table 4.3). Germination and plant growth trials were carried out with high volume loadings of hydrochar. While an increase in hydrochar concentration resulted in a decrease in plant germination and health, there was still significant growth for samples with loadings up to 30 vol%. Therefore, hydrochar did not completely inhibit germination and could be considered for land application as a method of carbon sequestration.

In most cases, regression models were found for the HTC product yields and their compositions for each of the feedstocks. These confirmed that hydrochar yield decreased but its carbon content increased with an increase in HTC temperature. The regression models were plotted and compared with the experimental data showing a good fit in most cases (Figures 4.17, 4.18 and 4.19). When compared with generic regression models for mass yield from literature, the models developed in this work gave better estimates (Figure 4.22). However, developing regression models based on parameters on a dry ash-free basis show promise as giving better predictions. Details of the experimental results are included in the Appendix, Table A.1 and Table A.2. The modelling results from this work were applied to the HTC process model in the following Chapter 5 Process Model. There, the optimum conditions in terms of a trade-off between product quality and plant costs are explored.

5 HTC Plant Model

5.1 Introduction

This chapter details the process modelling results based on the experimental results of the three types of digestate. This model explores the predicted hydrochar production, energy use, and the capital and operating expenditure of an industrial-scale HTC plant. To study the plant performance, the following work is presented:

- An overview of the plant design and typical industrial operating conditions that were used as defaults.
- A mass and energy balance of the plant at default conditions, including a consideration of the Greenhouse Gas emissions related to the energy use.
- The capital expenditure (CAPEX) and annual operating expenditure (OPEX) of the plant and each of the unit operations at default conditions.
- Validation of the process model using data from an industrial-scale facility as well as a comparison with literature results.
- The variation in results over a range of operating conditions and how the plant can be optimised for lowest cost or highest hydrochar energy content production.
- Consideration of the plant capacity and how economies of scale affect the costs.
- A brief comparison of the plant performance with lignocellulosic feedstocks compared to digestate.

The results here are from the process model and do not consider the markets of either the feedstocks or products. Chapter 6 uses a case study to further investigate the techno-economics, market potential and overall feasibility for industrial-scale HTC facilities.

5.2 Plant Design

A process model of HTC and thermal post-treatment was developed through a series of unit operations. Figure 5.1 shows each of these and the product flow streams. The model has a number of variables including type of feedstock, annual throughput, feedstock moisture content, reaction temperature and reaction time. It is flexible and can be run either at a single design condition or over a range of conditions. While the three types of digestate tested experimentally are built into the model, it is possible to investigate the HTC of alternative feedstocks by defining the properties and regression models.

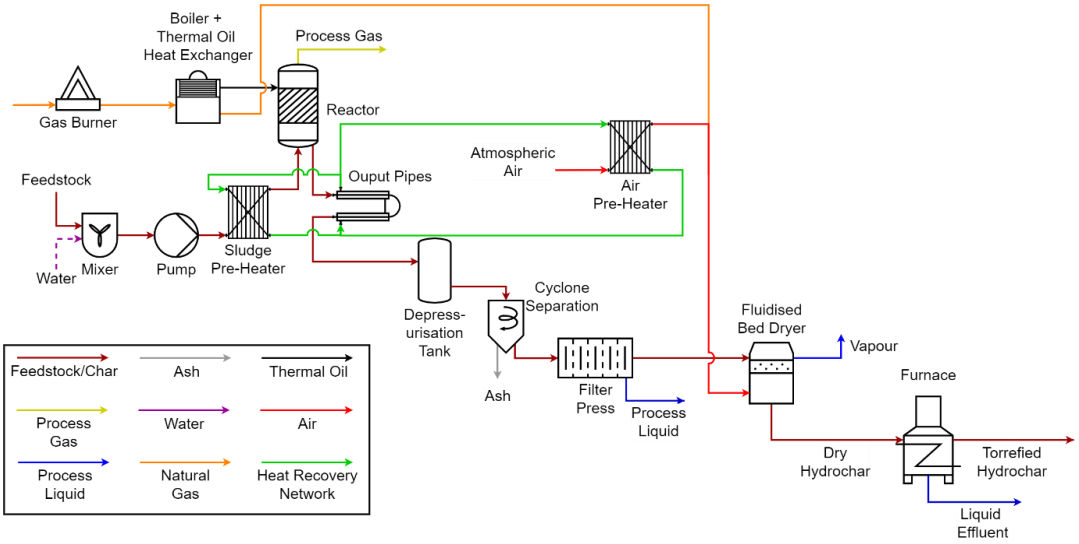


Figure 5.1: HTC process model unit operations.

The process model was based on the plans for an industrial-scale HTC facility. Due to the design of the HTC reactor, a modular system was used with each reactor able to hold up to 8 m^3 of wet feedstock. Therefore, with an operating time of 7,800 hours a year, each reactor could process roughly 21,000 tonnes of feedstock a year. While the HTC reactor had a fixed maximum size, other plant equipment such as the fluidised bed dryer and furnace were scalable.

5.2.1 Process model application

Using the MATLAB App Designer, a custom application was created to run and display the results of the model. The App was interactive and so the user was able to define the conditions of the plant and view how this affected the various outputs. The user interface is shown in Figure 5.2.

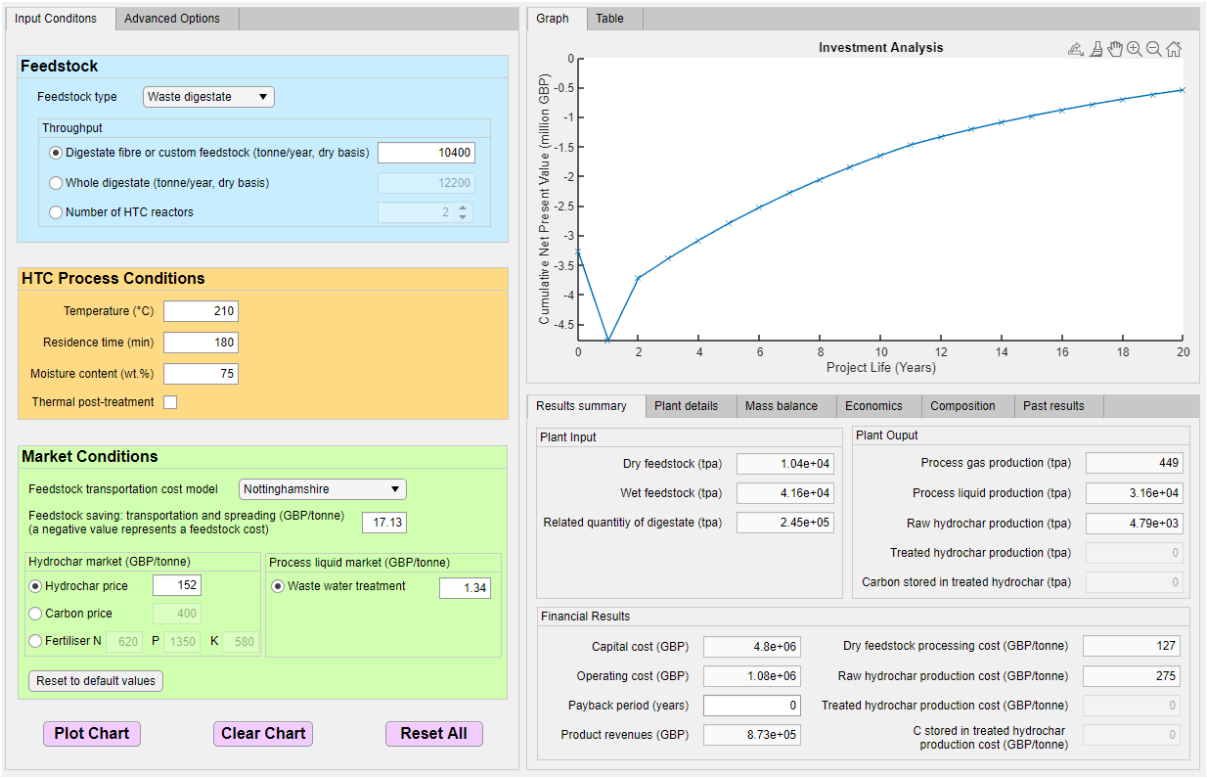


Figure 5.2: The user interface of the application created to run the process model.

The user defines the input conditions: feedstock (type and throughout), HTC process conditions (temperature, residence time, moisture content and presence of thermal post-treatment), and market conditions (location, feedstock saving and HTC product sale prices). When the user selects “Plot Chart”, the process model is run using the define input conditions. The process model consists of a number of functions, run in sequence. Details of these functions are shown in Figure A.1 in the Appendix. The rationale and calculations for each unit operation of the plant are given in Chapter 3 Methodology Section 3.4 Process Model.

The results of the process model are shown on the right-hand side of the app. The Investment Analysis is plotted showing the cumulative NPV over a 20-year timeline. The key results such as hydrochar produced, CAPEX and OPEX are displayed below in the Results Summary. There are a series of further tabs showing a more detailed breakdown of the results.

Using this app, the user is able to easily compare the results of different input conditions by plotting multiple Investment Analyses to the same figure and storing past results in a designated tab. This allows them to explore the technical and financial impact of different operating and/or market conditions. The app is fully functional and the app and process model source codes are available for further development.

5.2.2 Default conditions

Default HTC conditions were set to reflect common industrial operating conditions for a 2-reactor system [98], shown in Table 5.1. Digestate fibre with a moisture content of 75 wt.% was used, and therefore no make-up water was required to raise the moisture content of the feedstock on site. An investigation into the effect of economies of scale is included in Section 5.7.

Table 5.1: Default process model conditions.

	Variable	Default value
Feedstock	Source (waste, farm, crop)	Waste-based
	Type (whole, fibre)	Digestate fibre
	Annual throughput	41,600 tonnes/year
Reaction conditions	Temperature	210 °C
	Time	180 minutes
	Feedstock moisture content	75 wt.%
Products	Hydrochar moisture content	15 wt.% (based on industrial practice)
	Thermal post-treatment on/off	on
Temperature	Outside air	9.5 °C
	Pre-heat	155 °C
	Feedcake	20 °C

HTC produces a char product, but it has been shown that the use of raw hydrochar has negative effects when applied to agricultural soil. Therefore, results from a study using further thermal treatment to remove organic phytotoxic compounds were added to the model [86]. Literature shows that the optimum temperature for eliminating phytotoxicity but maintaining the highest solid recovery was at 275 °C. This gave a mass yield of hydrochar of 73.5% on a dry basis. It has been assumed that this thermal post-treatment densified the carbon content of the hydrochar by a factor of 1.06 [87].

5.3 Performance

The performance of the process model for each of the digestate feedstocks is shown in Table 5.2. Using the regression models developed in Chapter 4 HTC Test Programme Section 4.4, the mass yields of the hydrochar, process water and process gas were estimated. Note that the process water in this sense is the recovered liquid fraction from the filter press consisting of the majority of the process liquid produced in HTC along with the water content of the digestate fibre.

These results reflect the findings in Chapter 4 HTC Test Programme. The waste-based digestate produced hydrochar with a lower carbon content and higher ash content relative to the other types of digestate. The crop digestate produced more hydrochar in the process model due to its higher solid hydrochar yield, as found experimentally. This led to a higher thermal energy demand, particularly in the fluidised bed dryer and furnace, due to the larger quantity of hydrochar needing treatment. There was also a larger electricity demand for the crop digestate, again due to the increased throughput of the filter press and fluidised bed dryer. These increases in energy demand were reflected in the increased CO_2e emissions for the crop digestate.

Table 5.2: Mass balance of the HTC plant for the different feedstocks under default conditions: a HTC plant treating 41,600 tonnes of digestate fibre annually with a moisture content of 75 wt.% at 210 °C for 180 minutes.

	Waste digestate	**Farm digestate**	**Crop digestate**
Digestate fibre input (wet basis, t/y)	41,600	41,600	41,600
Digestate fibre input (dry basis, t/y)	10,400	10,400	10,400
Hydrochar production (15 wt.% mois., t/y)	5,500	6,700	7,300
Hydrochar production (dry basis, t/y)	4,800	5,900	6,300
Hydrochar carbon content (wt.% dry)	31.3%	47.5%	51.8%
Hydrochar ash content (wt.% dry)	42.3%	25.1%	6.5%
Treated hydrochar production (t/y)	3,500	4,300	4,700
Process water production (t/y)	31,500	29,400	28,400
Process gas production (t/y)	400	500	400
Water lost in drying (t/y)	4,100	5,000	5,400
Electrical energy use (MWh/y)	800	900	1,000
CO_2e emissions from electrical energy (t/y)	210	240	250
Thermal energy use (MWh/y)	8,100	9,400	10,000
CO_2e emissions from thermal energy (t/y)	1,500	1,700	1,800

The flow of feedstock and products of the waste-based digestate in Table 5.2 are displayed in Figure 5.3. While the hydrochar yield on a dry ash-free basis was 39 wt.% for the waste-based digestate, and up to 64 wt.% for the crop-based digestate, the production of hydrochar as a fraction of the total feedstock input was much lower. This is important when considering the moisture content of the feedstock and how that relates to the size of the plant.

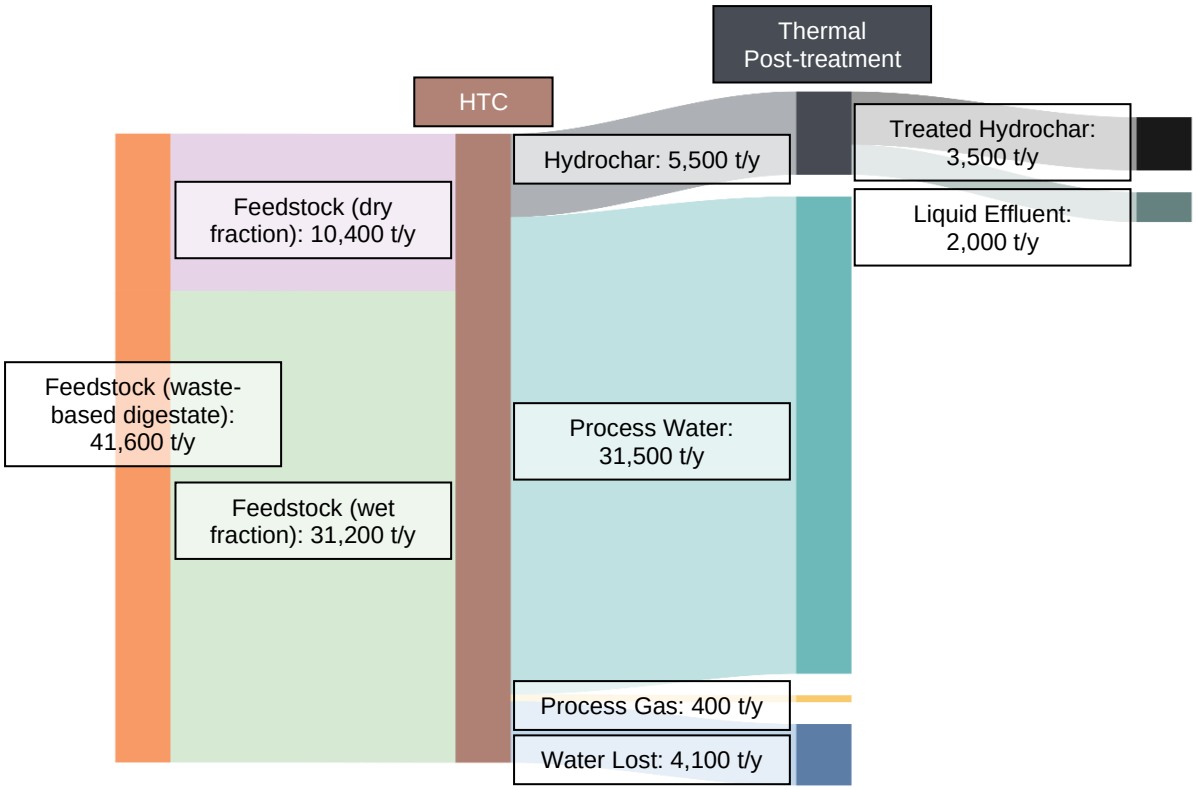


Figure 5.3: Sankey-diagram showing the mass balance of feedstock and products in the process.

5.3.1 Energy Use

Figure 5.4 shows the annual electrical and thermal energy use by the plant at the default conditions for the waste-based digestate. The thermal energy demand of the plant was around 10 times larger than the electrical energy requirement. The electrical energy demand mostly consisted of the air blower required by the fluidised bed dryer.

The process model uses a series of heat exchangers, details of which are given in Methodology Section 3.4.11. A steam boiler was modelled to supply all the excess heat not provided by the heat exchanger network. The heating of the air used for the fluidised bed dryer and feedstock heating accounted for 96% of the thermal energy demand. This can be attributed to the relatively large volume of wet feedstock requiring heating to reaction temperature, and the lower specific heat capacity of air used in the fluidised bed dryer. The feedstock passed through a double-pipe heat exchanger pre-heating system raising the temperature up to 155 °C (Figure 5.1). Heat losses from the reactor and heat generation from the exothermic reaction were accounted for and the extra heat was provided by the steam boiler. The air entering the fluidised bed dryer was raised from 9.5 °C, the average outside temperature in the UK [142], to the input temperature of 110 °C. The optional hydrochar post-treatment stage only processed the hydrochar and therefore was much smaller than the HTC equipment. The heat demand of this process was relatively low.

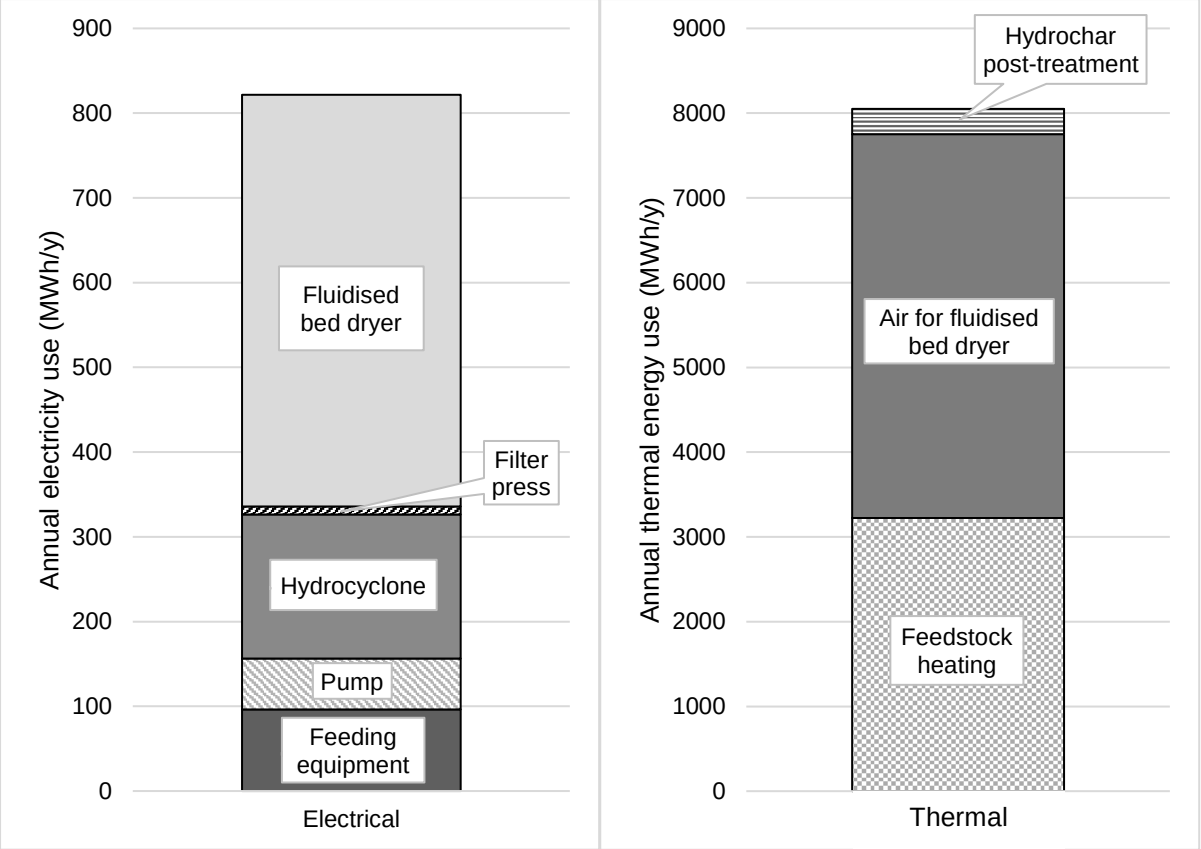


Figure 5.4: Average annual electrical and thermal energy use of the plant using default settings: a HTC plant treating 41,600 tonnes of waste-based digestate fibre annually with a moisture content of 75 wt.% at 210 °C for 180 minutes.

A series of heat exchangers were used in the process plant design, but in practice the thermal efficiency of the process could be improved further. If hydrochar post-treatment was to be installed, the liquid effluent, having 30–40% of the energy contained in the hydrochar, could be used for heating the HTC process [86]. As the HTC plant has been designed to process digestate, excess heat and biogas from the AD process could also be used in place of natural gas. This would reduce related greenhouse gas (GHG) emissions and the cost of fuel.

5.3.1.1 GHG Emissions

The GHG emissions related to these energy usages were estimated using conversion factors from 2019 [184]. The Department for Business, Energy & Industrial Strategy reported emissions of 0.1839 kg CO_2e/kWh for natural gas (gross) and 0.2556 kg CO_2e/kWh for electricity based on the average UK electricity mix. The transmission and distribution loss factor for UK electricity was 0.0217 kg CO_2e/kWh, bringing the total GHG emissions related to the UK electricity mix to 0.2773 kg CO_2e/kWh. Therefore, it was estimated that a HTC plant processing 41,600 tonnes of digestate fibre would release 1,490 tonnes of CO_2e from natural gas and 230 tonnes of CO_2e from electricity. If biogas were to be used in place of natural gas, the emissions would reduce to 0.00021 kg CO_2e/kWh, based on a biogas methane content of

65%. This would almost eliminate the release of GHG from thermal energy use down to 2 tonnes of CO_2e per year. These calculations do not account for GHG emissions from transportation, dewatering or distribution.

5.4 Costs

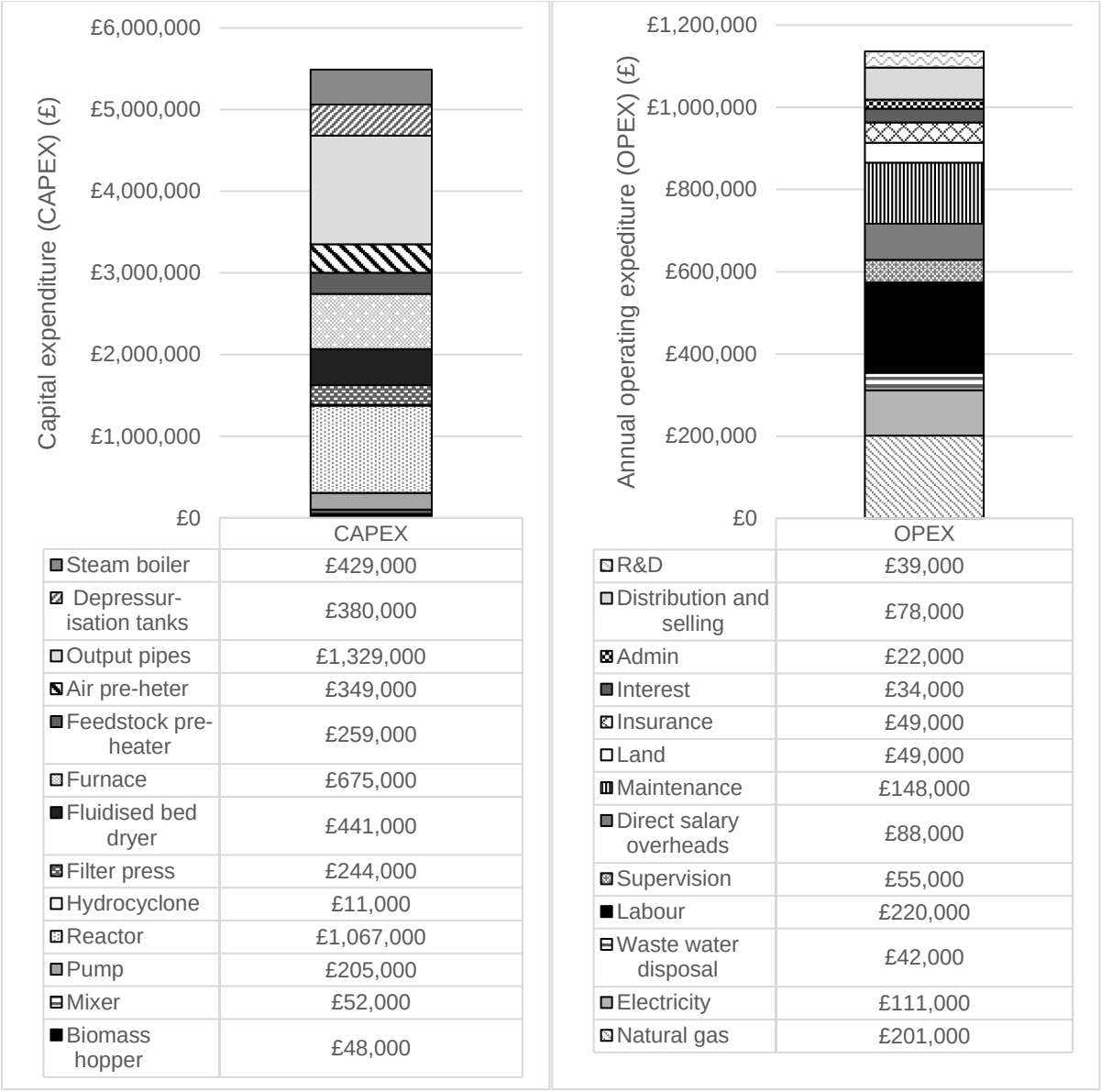
The CAPEX and OPEX for a plan of this size were calculated using engineering principles and publicly available data [152,154,155] (Chapter 3 Methodology, Section 3.5 Capital Expenditure and Section 3.6 Operating Expenditure). From these costs, the cost of producing hydrochar was estimated, as shown in Table 5.3. The fixed capital investment is the cost of the plant equipment. The total capital investment includes the working capital which consists of the upfront costs required before the plant is operational. These results show that the CAPEX and OPEX were similar for each of the feedstocks. The variation between them was due to the size of plant equipment and energy demand associated with separating and treating the differing quantities of hydrochar produced.

Each of these models were run using the default conditions, the feedstock input was held constant, and therefore the cost of processing 1 tonne of digestate was similar, varying from £34–36/tonne. However, the cost of producing 1 tonne of hydrochar was lower for the crop-based digestate at £239/tonne compared to £298/tonne for the waste-based digestate. This was due to the higher yield of hydrochar from crop-based digestate. When considering the market potential of the plant, the cost and availability of the feedstock can be more influential than the cost of the process itself. An investigation into this using an investment analysis and market predictions is given in Chapter 6.

Table 5.3: Cost analysis for a 2-reactor system for all three types of digestate.

	Waste digestate	**Farm digestate**	**Crop digestate**
Fixed capital investment (CAPEX) (£)	5,490,000	5,676,000	5,766,000
Total capital investment (£)	6,457,000	6,676,000	6,782,000
Operating expenditure (OPEX) (£/y)	1,155,000	1,209,000	1,236,000
Simple estimated annual cost (assuming CAPEX is split over 20 years) (£/y)	1,429,000	1,493,000	1,524,000
Digestate fibre feedstock input (t/y)	41,600	41,600	41,600
Digestate fibre processing cost (£/t)	34	36	37
Dry hydrochar production (t/y)	4,800	5,900	6,300
Hydrochar production cost (dry, £/t)	298	254	239
Treated hydrochar production (t/y)	3,500	4,300	4,700
Treated hydrochar production cost (£/t)	406	346	326

Figure 5.5 shows a breakdown of the CAPEX and OPEX costs for the waste-based digestate. The reactor and output pipes represented the highest plant equipment costs. Both of these had to accommodate the full quantity of wet throughput in the plant and operate at high temperatures resulting in higher costs. The labour costs and natural gas were the largest OPEX costs. Labour costs are location specific and the salary for plant operatives in the UK was taken as £22,873 [165]. There is also a large variation in the cost of natural gas globally and so these two costs could significantly alter the OPEX depending on location.



CAPEX	
Steam boiler	£429,000
Depressur-isation tanks	£380,000
Output pipes	£1,329,000
Air pre-heter	£349,000
Feedstock pre-heater	£259,000
Furnace	£675,000
Fluidised bed dryer	£441,000
Filter press	£244,000
Hydrocyclone	£11,000
Reactor	£1,067,000
Pump	£205,000
Mixer	£52,000
Biomass hopper	£48,000

OPEX	
R&D	£39,000
Distribution and selling	£78,000
Admin	£22,000
Interest	£34,000
Insurance	£49,000
Land	£49,000
Maintenance	£148,000
Direct salary overheads	£88,000
Supervision	£55,000
Labour	£220,000
Waste water disposal	£42,000
Electricity	£111,000
Natural gas	£201,000

Figure 5.5: The capital expenditure (CAPEX) and annual operating expenditure (OPEX) of the default plant: a HTC plant treating 41,600 tonnes of waste-based digestate fibre annually with a moisture content of 75 wt.% at 210 °C for 180 minutes.

5.5 Model Validation

The model produced in this work has been validated with data from an industrial facility, offering an improvement on what is currently available in the public domain. Figures 5.6, 5.7

and 5.8 show the energy use, CAPEX and OPEX for this model, an industrial facility, and a model from literature produced by Lucian et al. [135]. There is a lack of detailed HTC techno-economic models in literature and so the results of just one study was used. Each of the sources have been converted to a plant capacity basis for ease of comparison. While economies of scale affect the costs and energy usages of a plant, they have not been considered in this comparison work. All costs have been converted to GBP in 2019 using the same conversions as found in Chapter 3 Methodology Section 3.5. The hydrochar post-treatment has been omitted from the results from this model as it was not present in either the industrial facility or literature model.

Each of the models have a much larger thermal energy use than electrical energy use, representing between 88% and 94% of the total demand (Figure 5.6). The literature model seems to significantly underestimate the total energy use, coming in at only half the energy use of the industrial facility. The energy use of the model in this work is also lower than the industrial facility but to a lesser degree. Overall, this new model shows reasonable agreement with the energy usage of the industrial facility, given that the existing industrial facility is not optimised for a specific feedstock or throughput.

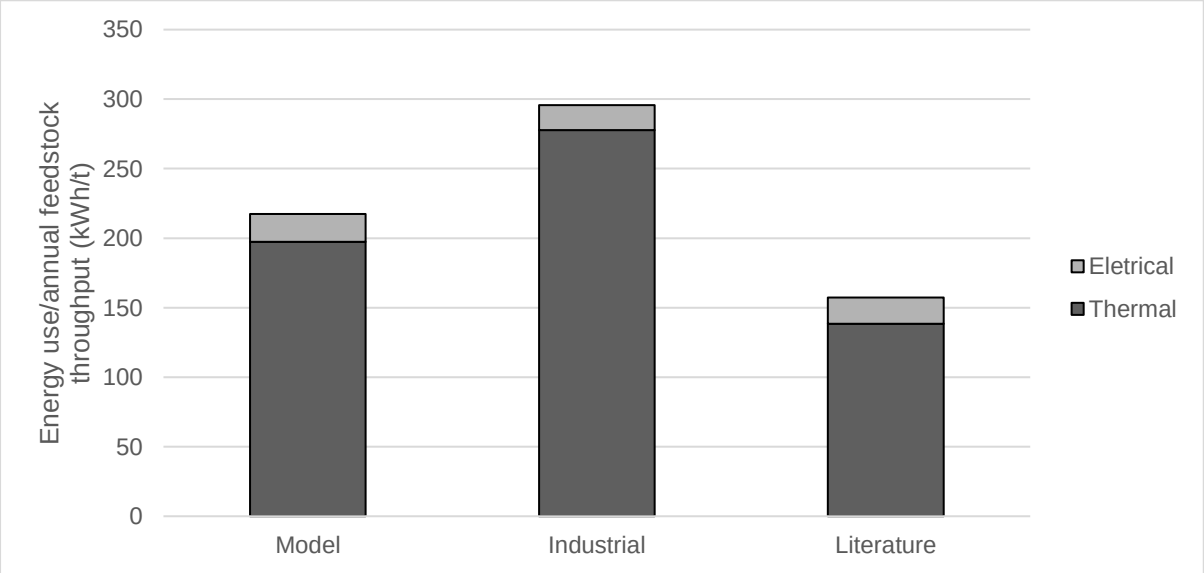


Figure 5.6: Model validation for energy use, compared to industrial data and a literature study [135].

Due to differences in reporting of plant equipment, each unit was categorised into feeding equipment (upstream of the HTC reactor), the reactor, separation (downstream of the HTC reactor), heat exchangers, and boiler. Accounting for the ±30% accuracy of the CAPEX estimation in this work, the model shows very good agreement with the industrial facility data. The total CAPEX values are similar and the contribution from each of the different types of plant equipment are also comparable. The literature model, however, shows a much lower CAPEX, suggesting there are under-estimates in costing all parts of the plant equipment.

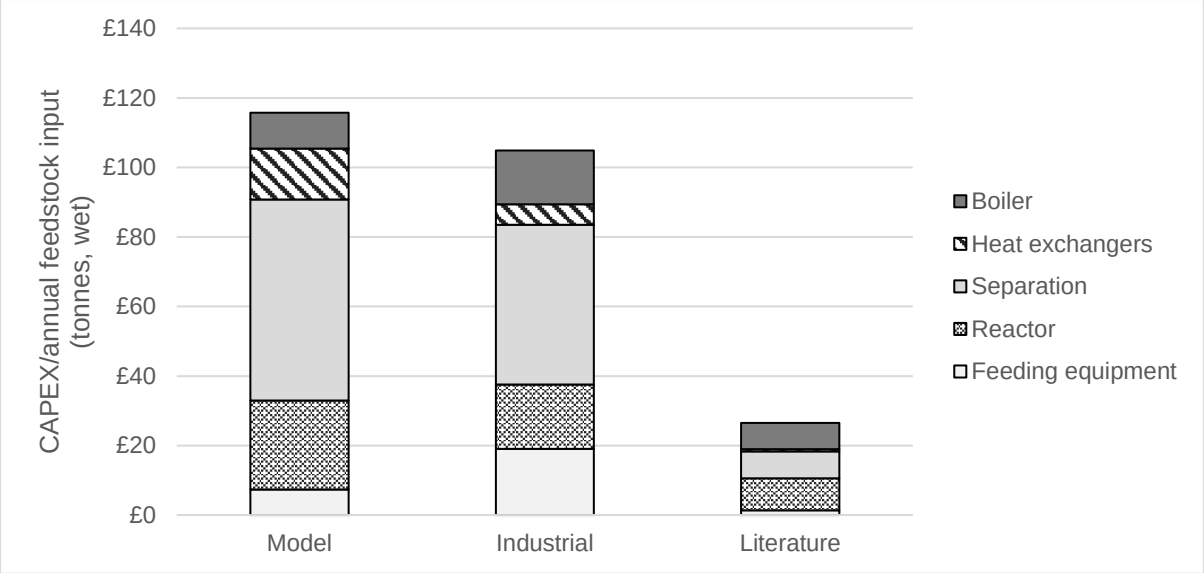


Figure 5.7: Model validation for CAPEX, compared to industrial data and a literature study [135].

The fractions making up the OPEX were also re-categorised to account for differences in reporting. The model in this work gave the largest OPEX value, with proportionally larger values for labour costs, maintenance and administration compared to the industrial facility. The labour costs in this work are based on UK wages which are on average higher than wages in Spain and Italy, the locations of the industrial facility and literature model, respectively [185]. The literature model gave a much lower OPEX value than the industrial facility. This can mostly be attributed to the lower thermal energy use estimate, and therefore lower cost of natural gas. It was also noted that the literature study only had 1 shift position with 4 operators per shift. It is recommended for a plant of that size to have at least 2 shift positions with 4.8 operators per shift [162]. Therefore, the overall labour costs in the literature study were deemed insufficient.

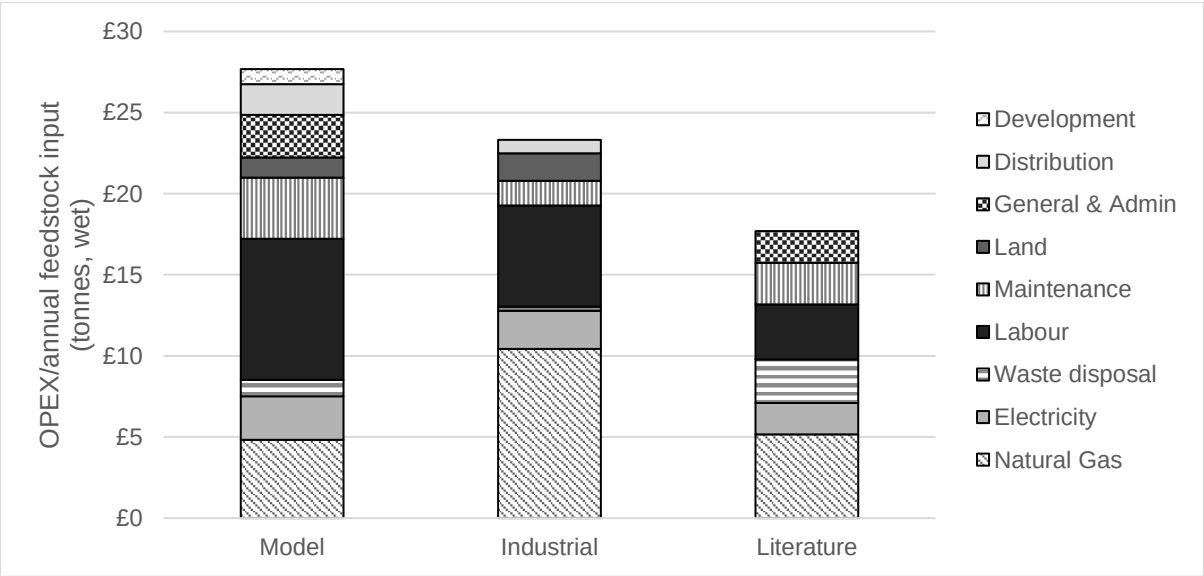


Figure 5.8: Model validation for OPEX, compared to industrial data and a literature study [135].

Overall, the model produced in this work showed good compliance with the energy use and cost data from an industrial facility. This supports the validity of using this model for further work and market predictions.

5.6 Optimisation

Much of the experimental work was looking at the effects that different operating conditions had on the product yields and properties. In summary, it was found that at higher temperatures and higher moisture contents, there was a lower yield of hydrochar but an increase in its carbon content. When considering operation at industrial scale, the process conditions are of interest in terms of product yields and quantities as well as costs.

Current industrial HTC reactors use the density of the reaction products to separate them from the feedstock in a stationary, non-agitated pressure vessel. This allows for continuous operation and a reduction in cost due to the minimisation of moving parts. In order for this system to function, the reaction mixture must remain in the reactor for a minimum residence time. The size of the reactor dictates the residence time. While residence times as low as 60 minutes were tested experimentally, an industrial size plant require longer residence times in practice. Therefore, when considering the results of the process model, the residence time was varied only between 120 and 180 minutes. This fits with the total processing time of several hours as reported for an industrial facility [98].

Additionally, when considering HTC on an industrial scale, the moisture content of the feedstock affects the logistics and size of plant equipment required. While it was found that having higher moisture contents increased the carbon content of hydrochar, in practice, lower values of moisture content are preferred. Therefore, it is likely that digestate fibre would be preferred in HTC, typically having a moisture content of 75 wt.% [20]. To illustrate this point, the default plant model treats 10,400 tonnes of dry feedstock, equivalent to 41,600 tonnes on a wet basis when the 75 wt.% moisture content is taken into account. If the same quantity of dry matter was treated but with a moisture content of 95 wt.%, typically as found with whole digestate, the HTC plant would need to process 218,300 tonnes of total feedstock a year. This would require 5 times the amount of reactor vessels, with all other plant equipment having to scale up accordingly. The CAPEX would increase from £5.5 million to £22.5 million. The annual OPEX would increase from just below £1.2 million to £4.4 million. Additionally, typical biomass feeding equipment is designed for feedstocks with lower moisture contents and so specialist units would need to be used for whole digestate. Using the cost estimate from WRAP, as detailed in Chapter 3 Methodology Section 3.4.2, dewatering 207,900 tonnes of whole digestate would cost in the region of £2 million to produce 41,600 tonnes of digestate fibre with a moisture content of 75 wt.%. Therefore, despite this extra cost, there is still an

overall cost saving associated with dewatering whole digestate to us digestate fibre in the HTC plant.

In light of these industrial considerations, the moisture content was set at 75 wt.%, the residence time ranged between 120 minutes and 180 minutes and the temperature was varied between 180 °C and 250 °C when assessing the changes in the plant model.

5.6.1 CAPEX

Figure 5.9 shows how the CAPEX changed with operating conditions. From Figure 5.9a, there are noticeable jumps in capital cost with an increase in residence time. After looking at the increase in capital cost for each of the unit operations, these were found to correlate with the increase in reactor cost. As displayed in Figure 5.5, the reactor cost represented 20% of the total CAPEX of the plant. As the residence time increased, the size of the reactor increased to accommodate the extra feedstock contained at any one time. This is confirmed in Figure 5.9b. The 4 distinct steps represent a unit reactor volume of $8m^3$, $9m^3$, $10m^3$ and $11m^3$ respectively, operating at a volume capacity of 65–75% [149]. Note that the total plant capacity was held at 41,600 tonnes/year for each of these runs, which resulted in a 2-reactor design. An important factor that has not been considered in this work is that as HTC is performed in a closed vessel, an increase in HTC temperature results in an increased pressure inside the reactor. The reactor would need to have thicker walls, thus increasing the reactor CAPEX at higher temperatures. As seen in Figure 5.9a, there were also dips in capital cost with an increase in HTC temperature. These were because of the combined effect of the output pipes, depressurisation tanks and steam boiler costs. The output pipes (Figure 5.9c) were double-pipe heat exchangers. At higher HTC temperatures, the area, and therefore cost, of the output pipes decreased as the slurry leaving the reaction was at a higher temperature. Therefore, a smaller area was required for heating up the water in the heat recovery line. Also, at shorter residence times, a smaller quantity of slurry had to pass through the output pipes, decreasing their size and cost. The cost of the depressurisation tanks (Figure 5.9d) also decreased with a decrease in residence time, due to the smaller volume required. At lower temperatures, the density of the slurry mixture was higher, decreasing the size of the depressurisation tanks. Finally, an increase in reaction temperature lead to an increase in steam boiler size (Figure 5.9e) due to the larger duty required. It was the combination of all these effects on the unit operations that led to the variation in CAPEX as seen in Figure 5.9a.

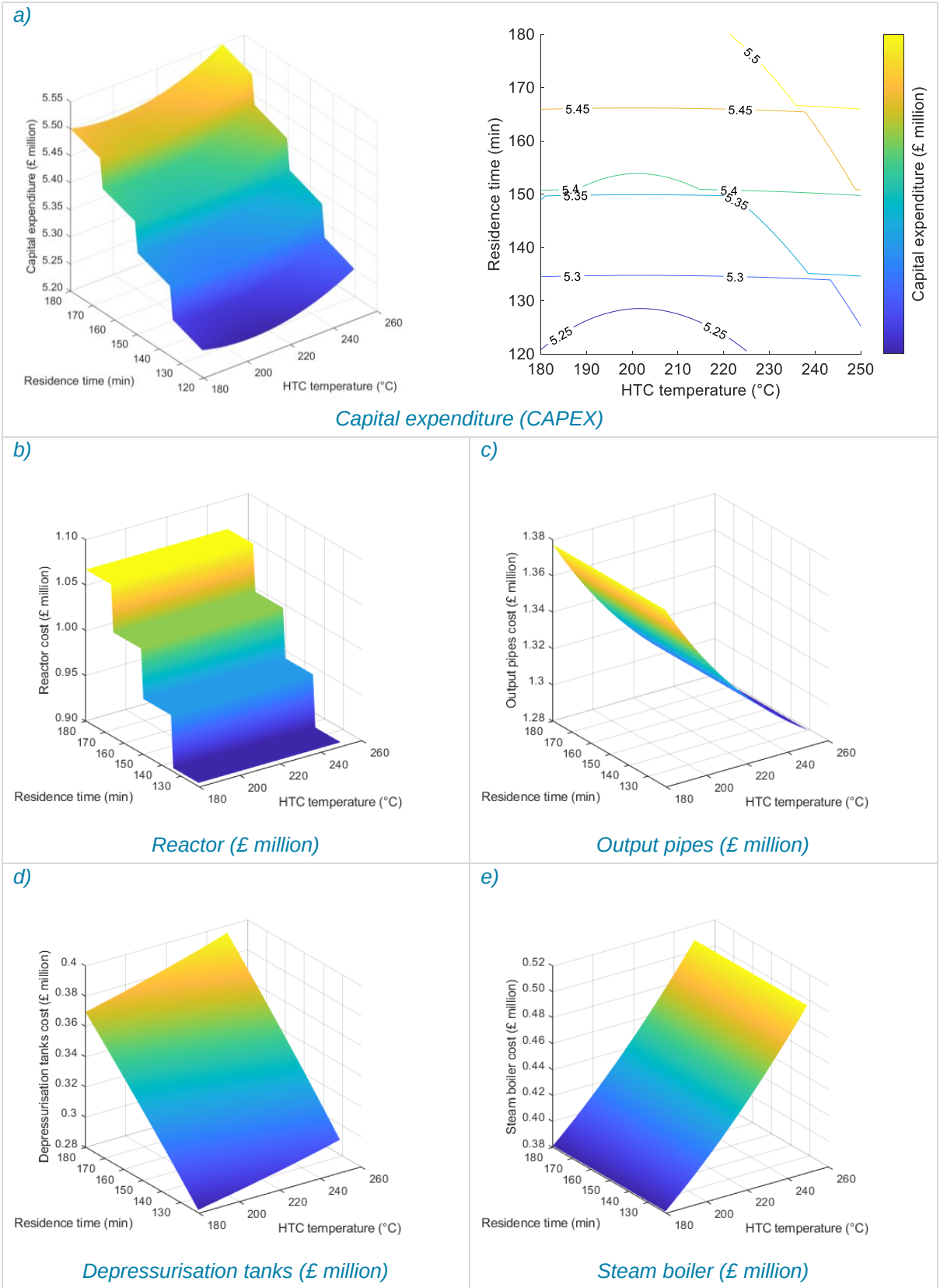


Figure 5.9: Change in a) total capital expenditure (CAPEX), b) the reactor cost, c) output pipes cost, d) depressurisation tanks cost, and e) steam boiler cost with a change in operating conditions.

5.6.2 OPEX

When considering the impact of operating conditions on the OPEX of the plant, a higher HTC temperature resulted in a higher cost. This is represented by the increase in scale in Figure 5.10a. The main component for this was the increase in the annual thermal energy requirement, and thus natural gas. The GHG emissions from thermal energy would follow the same trend. The annual electrical energy use was not greatly affected by a change in the process conditions, only differing by £3,000/year over the full range of conditions studied. The steps seen with an increase in residence time in Figure 5.10a are related to the increase in CAPEX. They match the increase in size and cost of the reactor (Figure 5.9b). As stated in the Methodology Section 3.6, the maintenance, land, insurance, and interest payments are derived from the CAPEX. Therefore, an increase in CAPEX leads to an increase in OPEX.

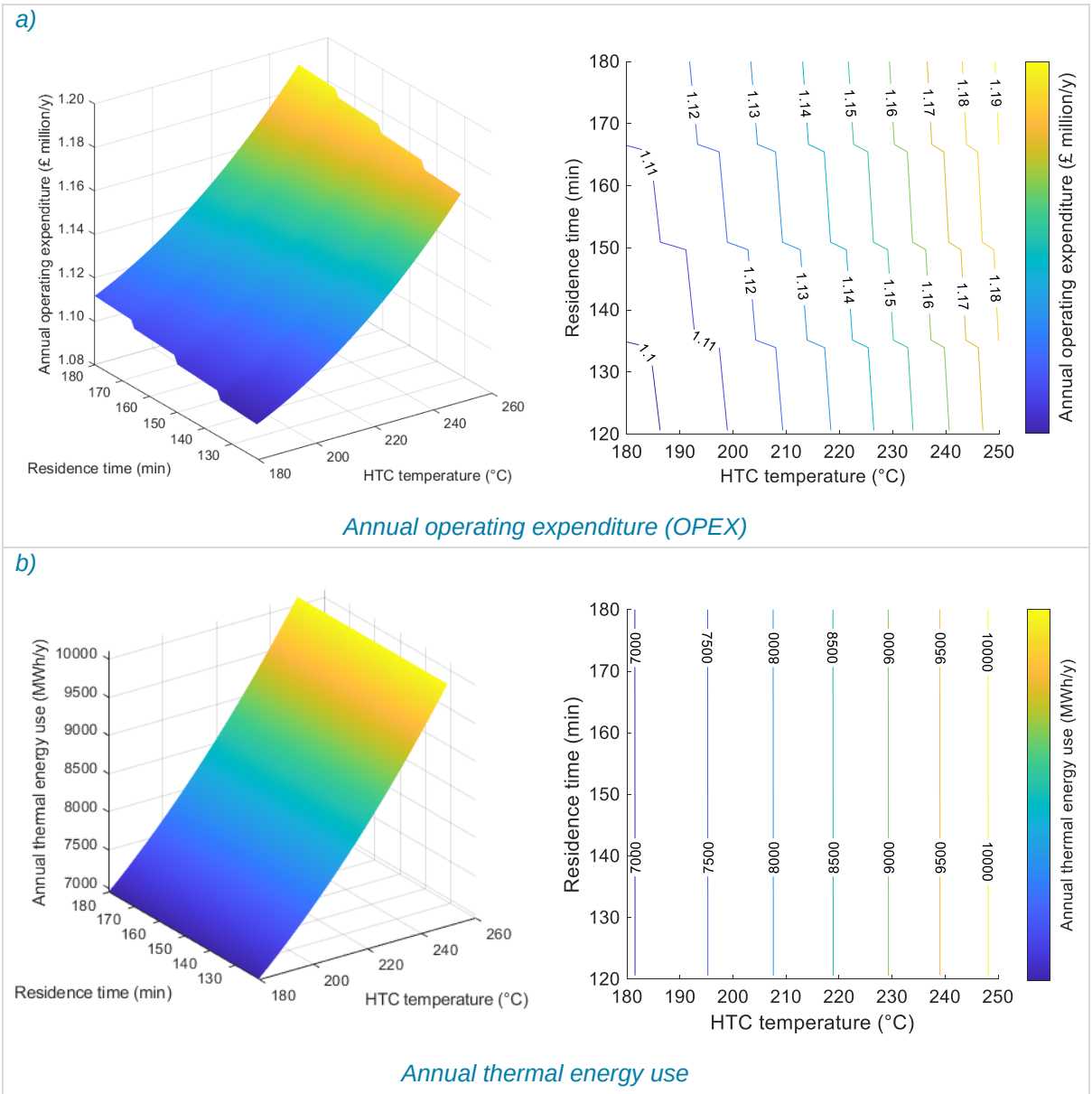


Figure 5.10: The change in a) OPEX and b) thermal energy usage with HTC temperature and residence time.

5.6.3 Processing Cost

For all the operating conditions investigated, the capacity of the plant remained constant and therefore the same quantity of digestate fibre was processed. The cost of processing 1 tonne of digestate fibre was a combined effect of the increase in CAPEX and OPEX with an increase in process condition severity as seen in Figure 5.11.

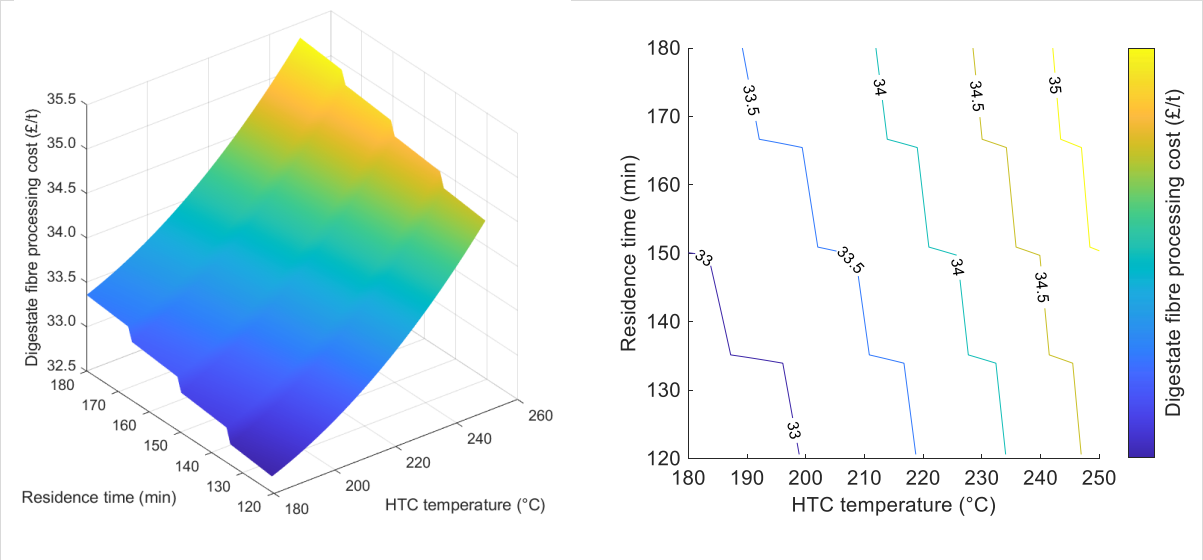


Figure 5.11: The change in digestate fibre processing cost with HTC temperature and residence time.

5.6.4 Production Cost

The effect of operating conditions on the yield and properties of hydrochar was reported in Chapter 4. As the regression equations developed in this work were fed into the process model, the overall conclusions remain true: an increase in HTC temperature decreased the yield of hydrochar but increased its carbon content. Therefore, the combined effect of an increase in CAPEX and OPEX and decrease in hydrochar yield led to an increased hydrochar production cost at higher HTC temperatures and longer residence times. However, when considering the optimisation of product properties, it is important to note the market price of the product. Although the cost of production of 1 tonne of hydrochar is higher at higher HTC temperatures, it could be offset by the increase in sale price due to its greater energy content. A fuller evaluation and consideration of market potential is given in Chapter 6.

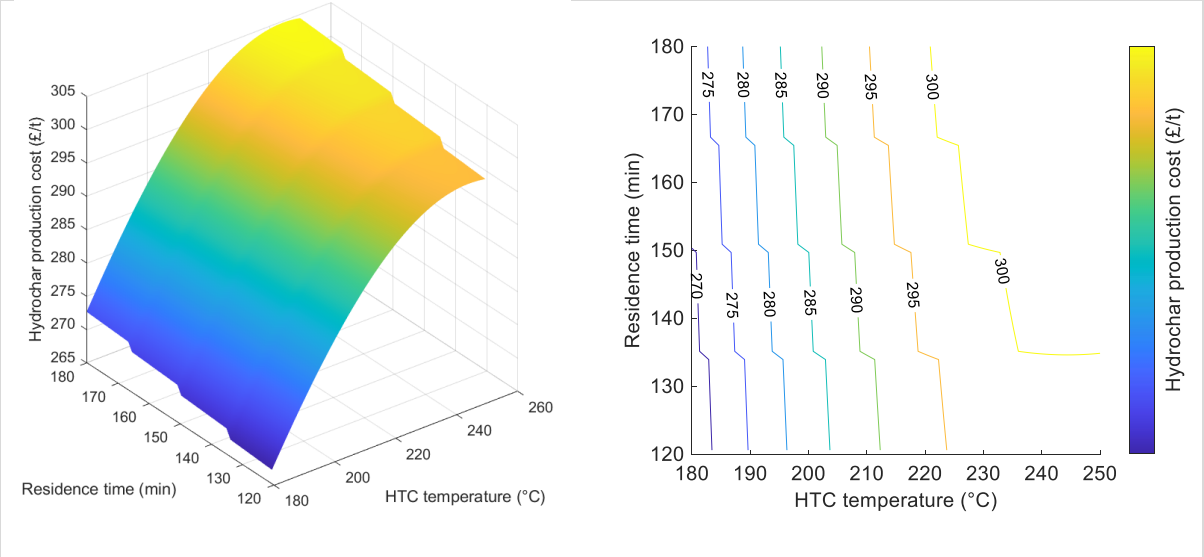


Figure 5.12: The change in hydrochar production cost with HTC temperature and residence time.

5.6.5 Processing conditions

Using the experimental and process model results, a simple optimisation for cost and hydrochar quality was performed. A comparison with the default conditions is shown in Table 5.4. The processing conditions resulting in the lowest cost and maximum hydrochar production were a HTC temperature of 180 °C and a 120 minutes residence time. Alternatively, a HTC temperature of 250 °C and residence time of 180 minutes resulted in hydrochar with the highest energy content. However, in practice, design and market considerations would need to be taken for each scenario. For example, some feedstocks are better processed at higher temperatures, rather than 180 °C, particularly those containing contaminants. As previously mentioned, the higher market price of hydrochar with a higher energy content would need to be compared to the increase in HTC production cost.

Table 5.4: Optimum conditions for cost and hydrochar quality compared to default values: a HTC plant treating 41,600 tonnes of waste-based digestate fibre annually with a moisture content of 75 wt.% at 210°C for 180 minutes.

	Default conditions	Minimum cost & Maximum hydrochar production	Highest hydrochar carbon content
Temperature (°C)	210	180	250
Residence time (min)	180	120	180
Feedstock input (wet basis, t/y)	41,600	41,600	41,600
Hydrochar production (dry basis, t/y)	4,800	5,100	4,800
Treated hydrochar production (t/y)	3,500	3,700	3,500
Process liquid production (t/y)	31,500	31,200	31,100
Process gas production (t/y)	400	200	800
CAPEX (£ million)	5.49	5.26	5.55
OPEX (£ million/y)	1.15	1.11	1.21
Digestate fibre processing cost (£/t)	34	33	36
Hydrochar production cost (£/t)	298	271	308
Thermal energy use (MWh/y)	8,100	6,900	10,100
Electrical energy use (MWh/y)	800	800	800
GHG (tonnes CO2e/y)	1,710	1,490	2,070

5.7 Plant Capacity

The default plant throughput was set to 41,600 tonnes of wet waste per year. Due to the flexible nature of the plant model, its capacity was altered to investigate the effect of economies of scale. The model was run, varying the input from 13,000 tonnes per year (a 1-reactor system) to 415,000 tonnes per year (a 20-reactor system). As discussed in Chapter 3 Methodology, the plant model consisted of both modular and scalable unit operations. The reactors were designed to hold up to 8m^3 of wet feedstock at any one point.

Figures 5.13a and 5.13b show the increase in the CAPEX and OPEX. As expected, an increase in plant capacity led to higher costs. Each step increase in cost represents an increase in the number of reactors required, between 1 and 20. It was shown in Section 5.4 Costs that the reactor cost represented a large fraction of the total CAPEX. Despite the increase in CAPEX and OPEX, an increase in plant capacity led to a decrease in digestate fibre processing cost and hydrochar production cost, as shown in Figures 5.13c and 5.13d. This highlights the benefit of economies of scale for the HTC plant. For each number of reactors, there was a local minimum in cost. The default plant conditions used in this chapter

are based on the local minimum processing cost for a 2-reactor system, as indicated by the red cross.

While it can be seen that an increase in plant capacity reduced the cost of the HTC plant per tonne of feedstock processed and hydrochar produced, transportation costs have not been considered in this analysis. Digestate fibre is produced at AD plants of varying size throughout the UK. Due to the high moisture content of digestate, there is a large cost associated with its transportation. One of the main drivers for its treatment with HTC is a reduction of the moisture content and therefore a reduction in transportation costs. A HTC plant of larger capacity would require digestate fibre from multiple AD plants as feedstock, incurring higher overall transportation costs. Therefore, it is important to weigh up the savings from economies of scale with the increasing cost of feedstock transportation. This is assessed in Chapter 6.

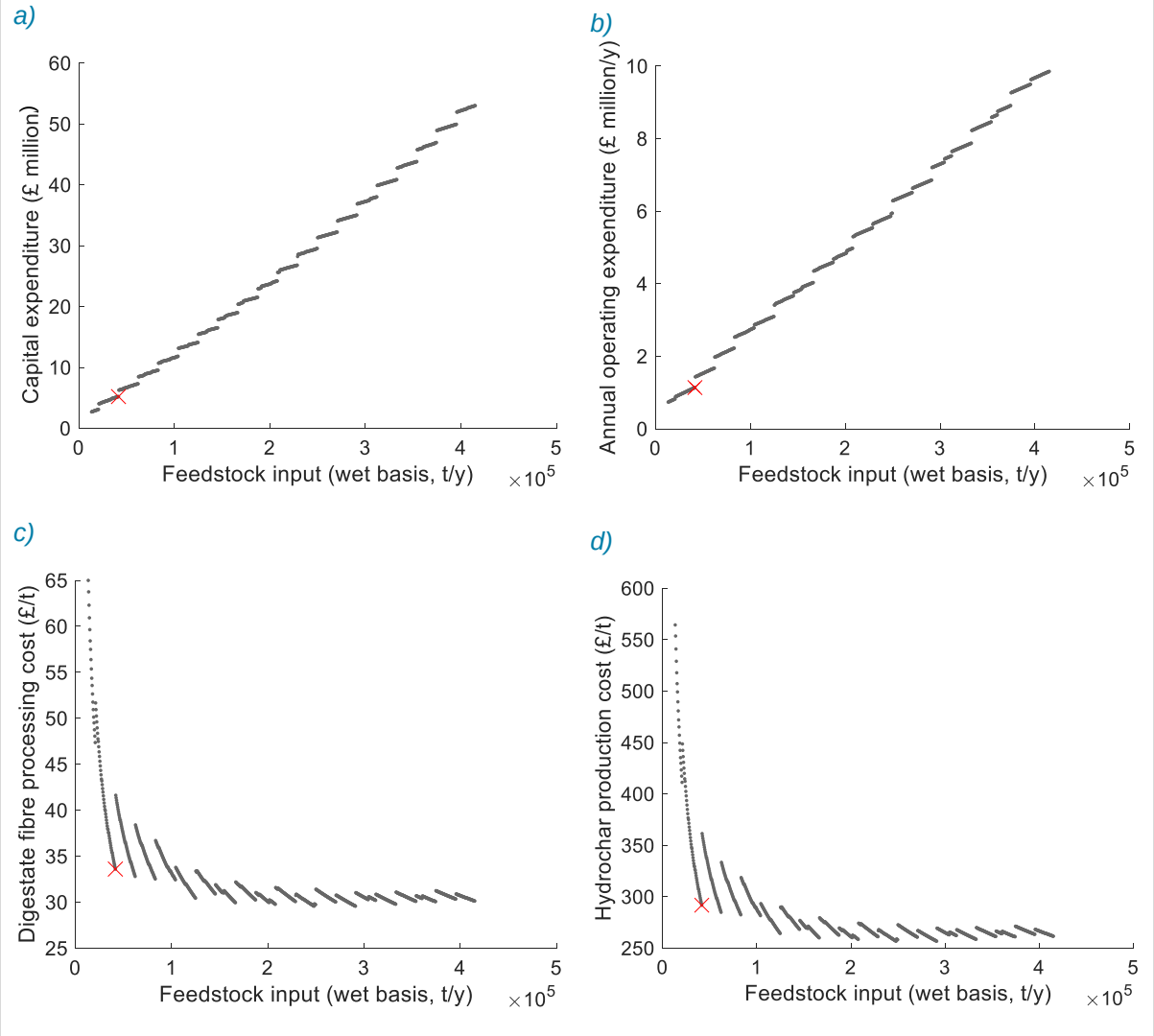


Figure 5.13: Change in CAPEX, OPEX, digestate fibre processing cost and hydrochar production cost with an increase in plant capacity. Results of the 2-reactor default system are marked with a red cross.

5.8 Alternative Feedstocks

The process model developed in this work is flexible and can be used with multiple inputs, detailed in Table 5.1. While this study is focussed on the HTC of digestate, the model can use any feedstock as long as the properties and regression models are defined. Specifically, these include the elemental analysis (C, H and N), proximate analysis (ash, fixed carbon, and volatile matter) and regression models for product yields and hydrochar composition. Data for a variety of feedstocks is available in literature and could be used in the model.

To study the techno-economics of lignocellulosic feedstocks, results from miscanthus and wood pellets of mixed spruce and pine were used. This work was previously undertaken at the University of Nottingham using the same experimental setup and methodology [186]. Simple regression models were created from the results of these experiments using the same technique as detailed in Section 3.3 Statistical Analysis of Results, Chapter 3 Methodology.

Figure 5.14 shows the production and composition of hydrochar for each of the feedstocks. The miscanthus and wood pellets produced similar quantities of hydrochar to that of the crop-based digestate, which were all noticeably higher than for the waste-based digestate. The lower ash contents and relatively large carbon contents of the two lignocellulosic feedstocks are also apparent. This results in the higher energy contents of 22.6 MJ/kg and 22.0 MJ/kg for the miscanthus and wood pellets hydrochars respectively, compared to 11.7 MJ/kg and 18.2 MJ/kg for the waste-based and crop-based digestate hydrochars, respectively. HTC of the two lignocellulosic feedstocks produced larger volumes and higher quality hydrochar products.

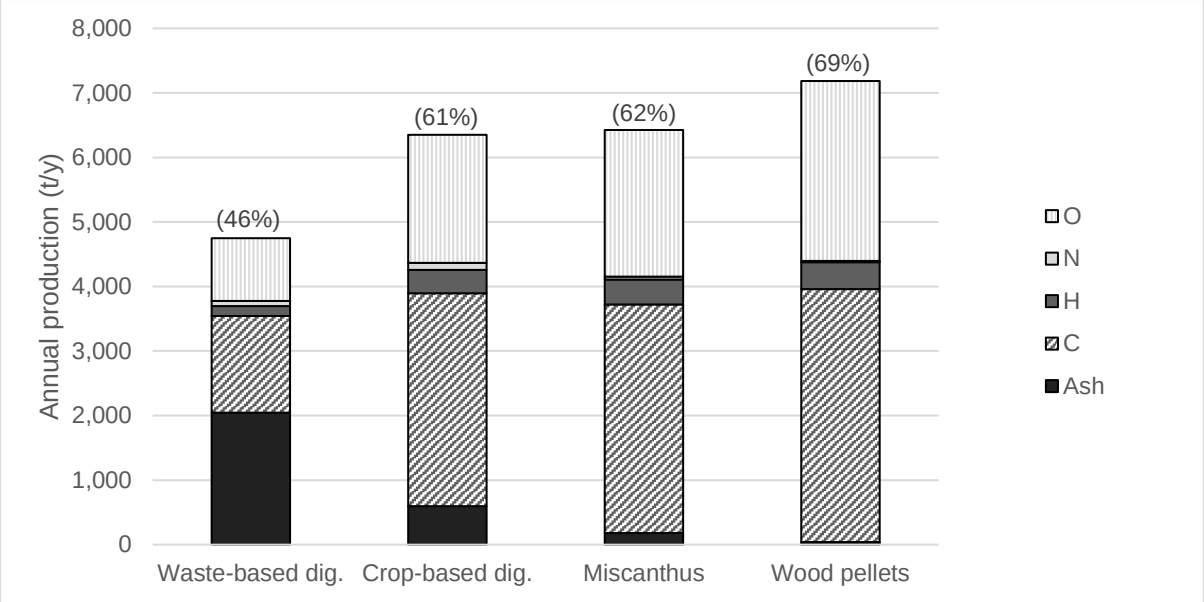


Figure 5.14: Hydrochar production and composition from waste-based digestate, crop-based digestate, miscanthus and wood pellets using the default process model conditions: a HTC plant treating 41,600 tonnes of feedstock annually with a moisture content of 75 wt.% at 210 °C for 180 minutes. The yield on a dry ash-free basis is shown above each of the bars.

The costs of the HTC of digestate and lignocellulosic biomasses were also compared. Figure 5.15 shows the CAPEX, OPEX, feedstock processing cost and hydrochar production cost for each of the feedstocks. There are only minor differences in the CAPEX and OPEX between the feedstocks. The variation exists due to the higher quantities of hydrochar produced from the lignocellulosic biomasses and crop-based digestate, requiring larger separation and drying equipment. As the default plant conditions were used here, the feedstock processing cost did not change much. However, there was a difference in the cost of production of 1 tonne of hydrochar. The increased production of hydrochar for the miscanthus and wood pellets resulted in lower production costs as the CAPEX and OPEX of the plant was relatively unchanged. Hydrochar production costs of £238/tonne for miscanthus and £220/tonne for wood pellets, comparable to £239/tonne for crop-based digestate and down from £298/tonne for waste-based digestate.

Although these results suggest that the HTC of miscanthus and wood pellets provide a lower cost opportunity for HTC, feedstock purchase costs and savings have not yet been considered. Multiple literature studies have highlighted feedstock cost as the most important factor for the financial feasibility of HTC, as detailed in Section 2.5.2 of Chapter 2 Literature Review. Digestate typically has costs associated with its treatment and transportation. Technologies such as HTC can offer a cheaper treatment solution and so the business model is based on savings that can be made. Miscanthus and wood pellets, however, are valuable biofuels in their own right and the HTC plant operator would have to factor in feedstock costs

as part of the investment analysis. Potential feedstock savings and costs are explored in Chapter 6 as part of the investment and market analysis.

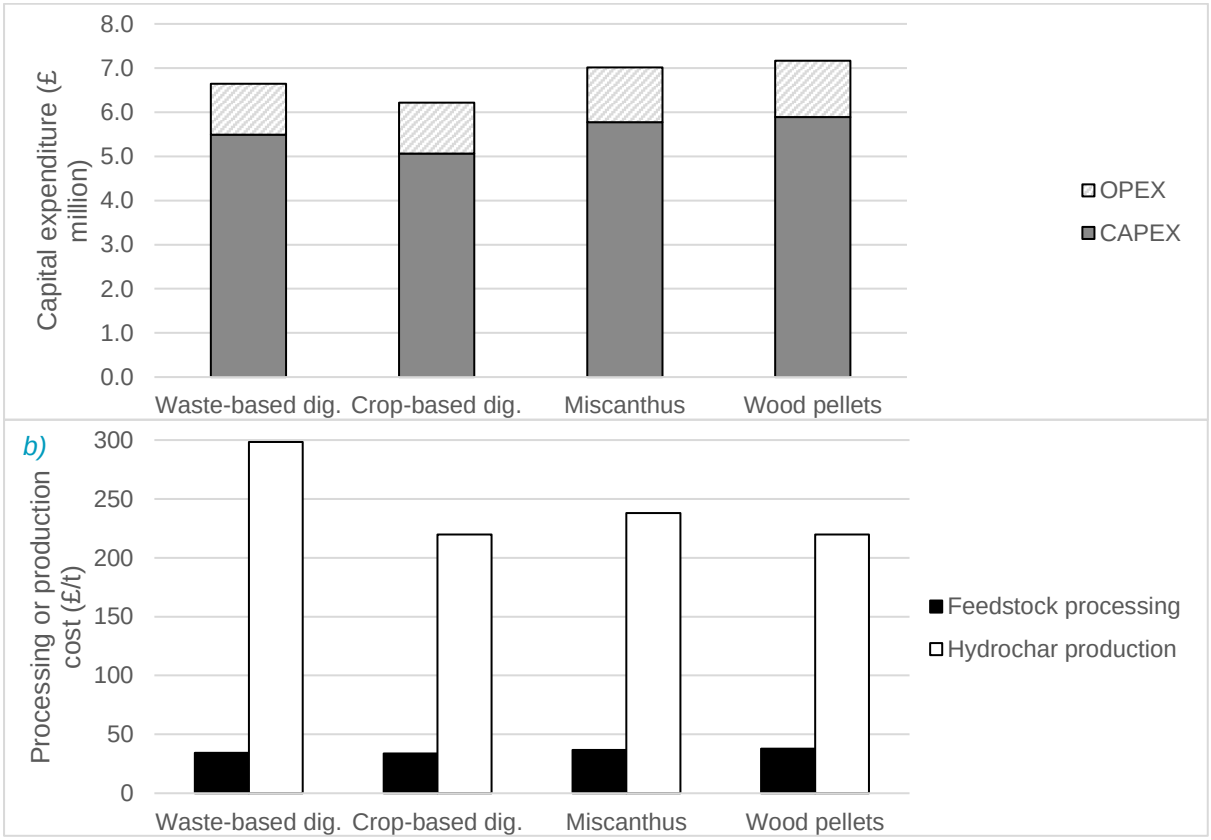


Figure 5.15: a) CAPEX & OPEX and b) digestate fibre processing cost & hydrochar production cost from waste-based digestate, crop-based digestate, miscanthus and wood pellets using the default process model conditions: a HTC plant treating 41,600 tonnes of feedstock annually with a moisture content of 75 wt.% at 210 °C for 180 minutes.

5.9 Conclusions

The majority of work and results in this chapter are based on the default conditions of a 2-reactor HTC plant, processing 41,600 tonnes of wet feedstock a year with a moisture content of 75 wt.%, operating at 210 °C and a residence time of 180 minutes. Depending on the type of digestate fibre, a HTC plant of this capacity was predicted to produce 4,800–6,300 tonnes of hydrochar, or 4,100–5,400 tonnes of treated hydrochar, annually. This required an electrical energy use of 800–1,000 MWh/y and a thermal energy use of 8,100–10,000 MWh/y, resulting in GHG emissions of between 1,700 and 2,100 tonnes of CO_2e annually. The thermal energy associated emissions could be greatly reduced if biogas were to be used in place of natural gas. The capital expenditure required to construct a HTC plant of this size came to £5.5–5.8 million and the annual operating costs were £1.2 million. The capital expenditure was dominated by the reactor and output pipes cost. Labour and natural gas costs were the largest contributors to operating costs. Taking into account the capacity and hydrochar production of

the plant, the cost of processing digestate fibre (with a moisture content of 75 wt.%) was estimated to be £34–37/tonne while the cost of hydrochar production came to £239–298/tonne.

The results of this model were validated with data from an operational industrial facility and were found to be in good agreement. Previous literature results, however, tended to give an under-estimate for energy use, CAPEX, and OPEX when compared to industry.

The process model was investigated under a range of operating conditions. It was found that operating at lower HTC temperatures and for shorter residence times lowered the cost of the process. However, operating at higher HTC temperatures gave hydrochar with a higher energy content. These results correspond to the experimental findings. The plant capacity was also varied, and it was found that there was a local minimum in terms of plant cost depending on the number of reactors used. The plant capacity used as default, 41,600 tonnes of wet waste annually, was the optimal condition for a 2-reactor system. An increase in plant size would lead to an increase in feedstock transportation cost which was not included in this analysis. A case-study concerning this is considered in Chapter 6. Finally, the plant performance for miscanthus and wood pellets, both lignocellulosic feedstocks, was investigated. Lignocellulosic feedstocks have higher hydrochar yields for HTC, resulting in larger quantities of hydrochar produced. With the CAPEX and OPEX varying very little between the feedstocks, the lignocellulosic feedstocks resulted in lower production costs per tonne of hydrochar, at £232/tonne for miscanthus and £214/tonne for wood pellets, compared to £298/tonne for waste-based digestate. However, the feedstock costs and savings were not considered in this analysis and therefore the economic feasibility was not fully understood. Details of this are included in Chapter 6.

Overall, this model successfully estimated the hydrochar production, energy use and costs for a HTC plant to a higher degree of accuracy than done before, as validated by industrial data. The model was flexible in nature and was used to explore a range of operating conditions looking to minimise cost or maximise hydrochar quality. Increasing plant capacity appeared to offer benefits on the basis of economies of scale, and lignocellulosic biomass feedstocks resulted in lower hydrochar production costs. However, both of these need to be studied further to understand the market potential and economic feasibility for the HTC process.

6 Case Study: Feasibility of HTC as a treatment for AD digestate

6.1 Introduction

This chapter presents a case study into the economic feasibility of HTC for the treatment of digestate. The feedstock and HTC products markets are coupled with the plant model developed in the process model. The following work consists of:

- The current status of digestate generation and management in the UK.
- A method for calculating the costs incurred in transporting digestate to the HTC plant location.
- An analysis of the dewatering of digestate, including its costs and benefits.
- A summary of the markets for HTC products.
- An investment analysis for the three types of digestate and the most common HTC product markets.
- The digestate savings and hydrochar sale prices required for certain payback periods.
- A sensitivity analysis based on the accuracy of the CAPEX estimates.
- A brief consideration of the economic potential of the HTC of lignocellulosic feedstocks.

The results are based on the experimental and process modelling work and use the same assumptions. As the market for many of the HTC products are not yet established, a range of values have been considered to give an overview of the potential of HTC.

6.2 AD in the UK

HTC is a relatively low-cost treatment for wet wastes, such as digestate, offering potential savings over their current management or disposal. It operates under wet conditions and therefore no drying pre-treatment is required. HTC also produces marketable products with hydrochar having potential as a solid fuel or soil amendment and HTC process liquid as a fertiliser. For the purposes of investigating the technical and economic feasibility of HTC, a case study was performed using digestate as a feedstock. Digestate is being generated at an increasing rate in the UK and there are many doubts over its future management and disposal strategies.

In 2019 over 12.5 million tonnes of feedstock were processed with anaerobic digestion (AD), generating 10.9 million tonnes of digestate by-product, equivalent to 500,000 tonnes on a dry basis [15,16]. Although there are delays in construction, as of April 2020 the number of AD plants rose to 579 with a further 331 under development [19]. Digestate composition depends on the feedstock used, especially the ash and carbon contents [187]. It has been estimated that there are between 133,000 and 240,000 tonnes of carbon contained in digestate annually,

shown in Table 6.1 [17,18]. AD capacity in the UK has increased over the last 15 years, supported by feed-in tariffs and renewable heat incentives [15], and is expected to continue to do so, especially for the treatment of food waste. Currently, 17% of AD plants process food waste, whether as a mono-substrate or co-digested with a variety of different feedstocks. WRAP estimated that in 2018, 9.5 million tonnes of food waste was generated within UK households, hospitality & food service, food manufacture, retail and wholesale sectors [172]. This had a value of over £19 billion and is associated with more than 25 million tonnes of greenhouse gas emissions. In excess of 1.3 million tonnes of food waste were fed directly to AD plants [188]. Looking ahead, there are governmental targets to reduce food waste generation over the next 10 years. If the Courtauld 2025 and SDG12.3 targets on UK food waste are met, overall food waste is projected to fall to 8.4 million tonnes in 2025 and 7.7 million tonnes in 2030 [172]. The UK government have identified that AD with digestate as a fertiliser material is environmentally better for treating unavoidable food waste than composting or other recovery options [13]. Assuming that 80% of food waste is directed to AD in 2030, that would represent 6.8 million tonnes, an increase of over 50% of the total AD capacity within 10 years.

However, current treatments of digestate are costly and legislation is likely to intensify. The standard practice for managing digestate is land spreading, with the criteria for this set out in the PAS110 [164]. Digestate is typically transported on a 50 mile round-trip from the AD plant to land [20]. At an average cost of £0.25/tonne, to transport digestate fibre with a moisture content of 75 wt.% one mile comes to a typical transportation cost of £12.50/tonne. The average cost of band spreading digestate fibre is £3.50/tonne. Therefore, the total cost of transporting and spreading dewatered digestate fibre comes to £16/tonne [21]. In addition to this, there are further regulations and associated costs if the digestate is to be spread to land designated as a Nitrate Vulnerable Zone (NVZ). Within NVZs, at least 5 months of digestate storage must be provided [173]. This can come at a high capital cost for the AD operator at £25–£50/m^3 (roughly £25–£50/tonne) of digestate stored [22,23]. Furthermore, with the introduction of the Reduction and Prevention of Agricultural Diffuse Pollution (England) Regulations 2018, digestate spreading is also regulated by the Environmental Agency. In practical terms, this results in a shorter spreading period for digestate and over 6 months storage is required in most situations. New storage requirements mean that AD operators are having to retrofit impermeable storage covers, often costing more than £50/m^3 to carry out. On top of this, there are concerns about the growing area of land designated as an NVZ. Draft legislation in Wales from April 2020 proposed to designate the whole of Wales as an NVZ, over 40 times the area currently covered [169]. The costs incurred depend on the location and scale of the AD plants. For farm-based AD plants, currently 92% of whole digestate is used

on site and the remainder is generally provided free of charge to off-site users. However, for commercial AD plants, 26% of the produced digestate was removed at cost to the operator in 2013. The issue is even more apparent for larger-scale industrial operators, where 44% of digestate was removed by off-site companies at a cost, 33% was supplied free of charge and 23% was disposed to sewers at a cost [24]. With increasing legislation and thus increasing costs expected, cheaper alternatives will be of interest to operators across the sector. A worst case scenario would be having to landfill digestate, with the average gate fee including landfill tax coming to £113/tonne [189,190]. The cost of incineration is not much lower at £89/tonne on average [190].

HTC of digestate has been shown to produce a solid hydrochar, recovering most of the carbon contained in the feedstock, as well as a nutrient-rich process liquid. Details of the process conditions, product characteristics and suitability in a range of markets are included in Chapter 4 HTC Test Programme. Table 6.1 provides a range of estimates for the quantity of carbon from digestate that can be stored in hydrochar. An advantage of HTC that is of particular interest for digestate treatment is its ability to process small quantities of contaminants, such as plastics. Plastics are a common contaminant found in food waste and are not broken down in the AD process. This means that food waste digestate often has to be mechanically processed, at a cost to the operator, in order to meet the PAS110 threshold required for land spreading [23]. The capital and operating costs of HTC were explored in Chapter 5 HTC Plant Model.

*Table 6.1: Potential carbon storage from digestate in the UK. All carbon contents are given on a dry basis. *experimental result for waste-based digestate, †experimental result for crop-based digestate.*

	Low	Mid	High
Feedstock processed by AD in the UK, May 2019 (t/y, wet) [15]		12,477,000	
Conversion rate [16]		87 wt.%	
Whole digestate production (t/y, wet)		10,855,000	
Whole digestate production (t/y, dry)		543,000	
Dewatering solids capture rate [20]		85%	
Digestate fibre production (t/y, dry)		461,000	
Carbon content of digestate fibre (dry)	29 wt.%*	34 wt.%†	52 wt.% [3]
Carbon contained in digestate fibre (t/y, dry)	133,000	157,000	240,000
Carbon recovery in hydrochar	51%*	71%	91%†
Carbon contained in hydrochar (t/y, dry)	68,000	123,000	219,000
Mass yield from thermal post-treatment [86]		73.5 wt.%	
Carbon densification after thermal post-treatment [87]		1.06	
Overall carbon recovery from the digestate to the treated hydrochar	40%	55%	71%
Potential carbon storage (t/y, dry)	53,000	87,000	170,000

6.2.1 HTC location and transport costs

When processing waste, its availability and location is fundamental when considering the plant scale. Digestate is produced throughout the UK, often at small-scale and in rural locations, although this could be set to change given the predicted increase in food waste digestion. One of the attractions of HTC for digestate is its potential as a cheaper alternative to transporting and spreading. However, a HTC plant processing larger quantities of digestate would have to source the feedstock from a greater area, incurring higher transportation costs. Therefore, two English counties were taken as case studies to analyse the trade-off between transportation distance and HTC plant size. Lincolnshire is the largest producer in the country, producing 603 ktonnes of digestate annually from 31 AD plants. Nottinghamshire is the English county with the highest density of digestate production, producing 360 ktonnes of digestate annually from 13 AD plants. This analysis assumes that dewatering down to a moisture content of 75 wt.% is carried out on-site by the AD operator at no cost to the HTC plant. Accounting for an 85% solid recovery during dewatering, this correlates to an annual production of digestate fibre of 61 ktonnes and 103 ktonnes in Nottinghamshire and Lincolnshire respectively.

Data for each of the AD facilities in both counties was collected [15]. A method was developed to find the optimum location for a HTC plant capable of treating all the digestate fibre produced in the county. The counties were split into a grid of 100 m^2 intervals. The cost of transporting the digestate from the existing AD plants within the county to each interval was estimated using a cost of £0.25 to transport 1 tonne of digestate fibre 1 km [21]. Once the point of lowest cost was found, shown in Figure 6.1, the coordinates were converted to a UK postcode. For Nottinghamshire, the best location for a HTC plant treating all the county's waste was NG14 5HJ (Figure 6.1.a); notably the location of the second largest digestate producer in the county and only a few kilometres away from the largest digestate producer. The same process was repeated for Lincolnshire and the best location was found as LN4 3QR, roughly in the centre of the county (Figure 6.1b).

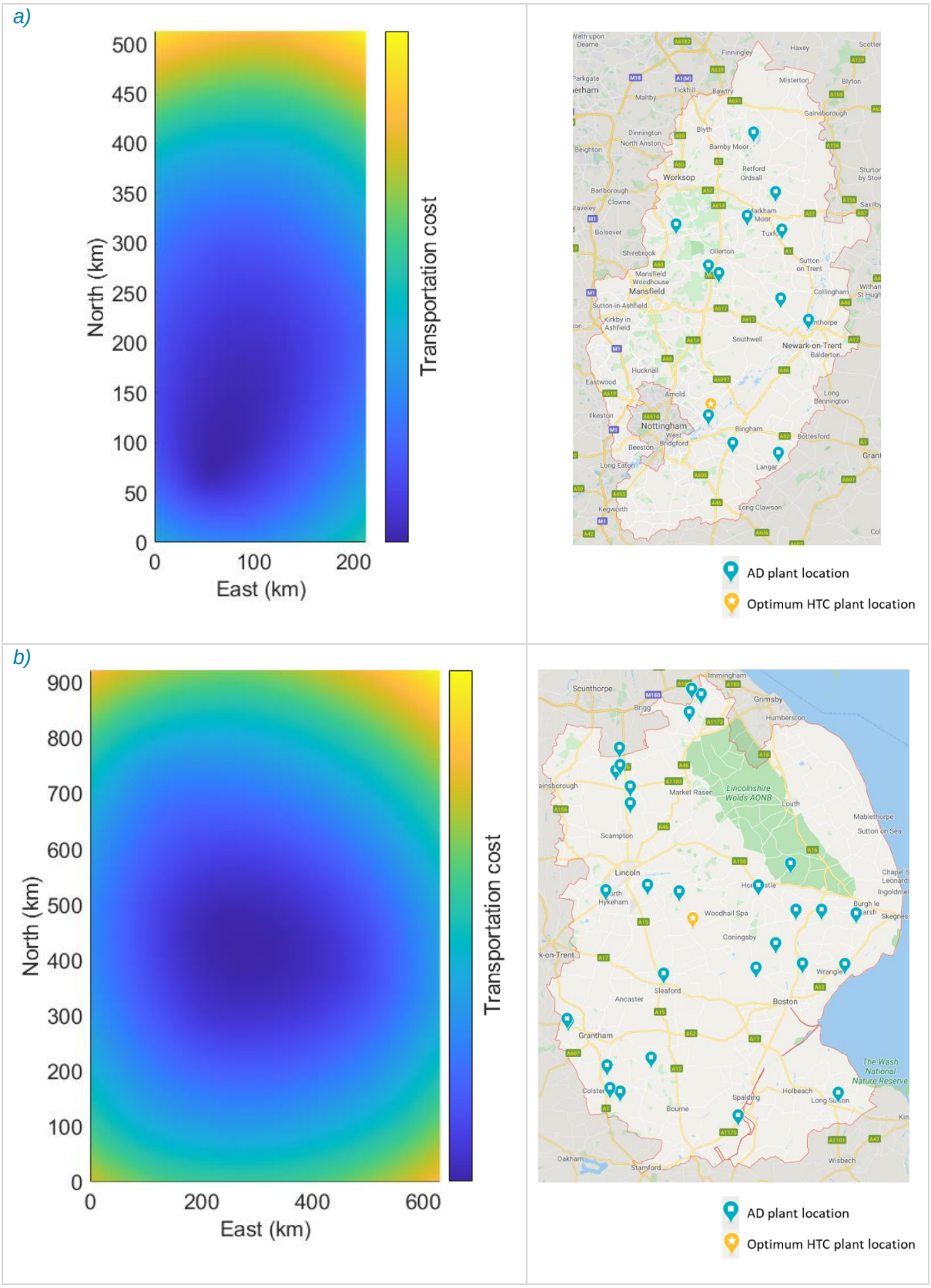


Figure 6.1: Map of a) Nottinghamshire and b) Lincolnshire with the location of each of the AD plants and the optimum HTC plant location. Map data ©2020 Google [191].

Once the postcodes of the optimum HTC plant locations had been found, driving distances from each of the AD plants were then measured for a more accurate calculation of transportation cost. The plants were ordered in distance from the optimum location and a cumulative sum of digestate fibre to be transported to the plant and the resulting transportation cost were calculated. The results of this are shown in Figure 6.2. There was a similar relationship for both counties, with a good fit to a second order polynomial relationship.

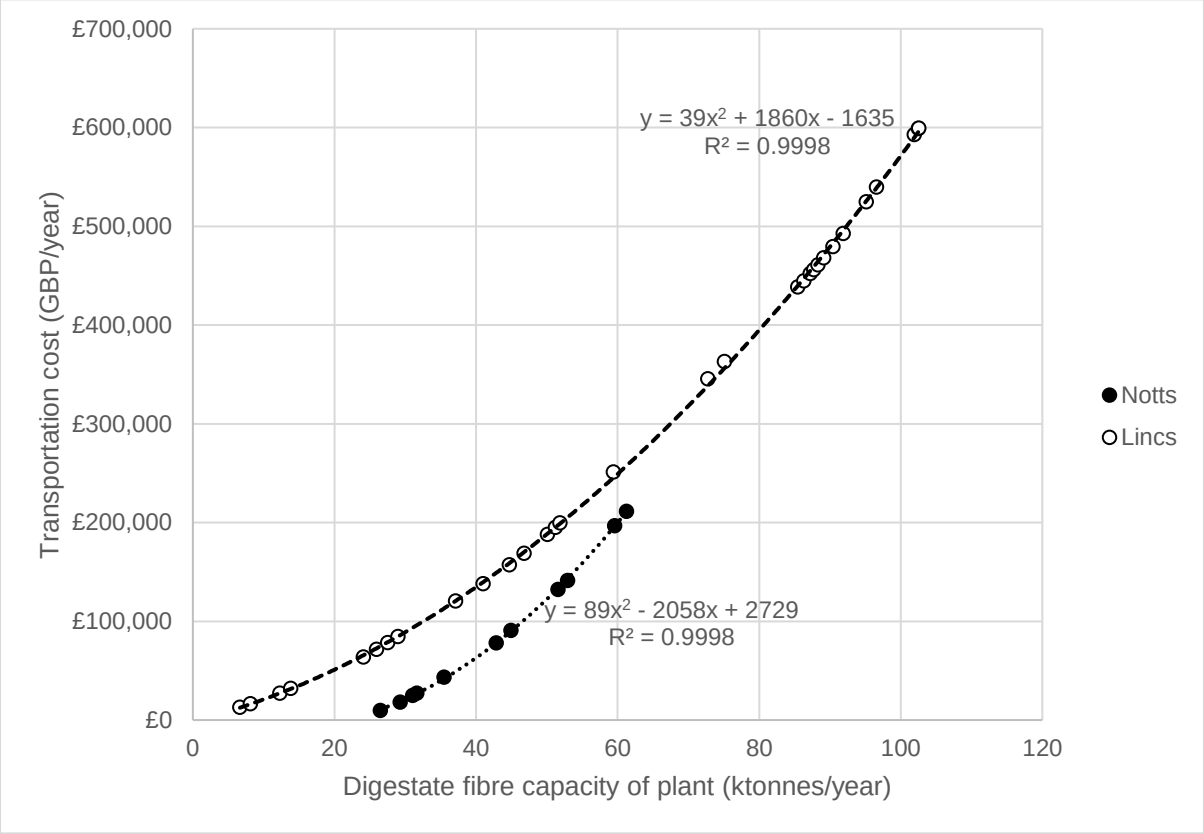


Figure 6.2: Increase in annual transportation costs with an increase in HTC plant capacity for both Nottinghamshire (Notts) and Lincolnshire (Lincs).

These polynomial relationships were then incorporated into the process model and the digestate fibre processing cost was calculated under the three scenarios, shown in Figure 6.3: excluding transportation cost, modelling transportation cost in Nottinghamshire and modelling transportation cost in Lincolnshire. As discussed in Chapter 5 HTC Plant Model, Section 5.7 Plant Capacity, each step increase in processing cost represents an increase in the number of reactors of the HTC plant. Nottinghamshire produces a lower quantity of digestate fibre annually compared to Lincolnshire. For Nottinghamshire, the lowest cost per tonne of digestate fibre was found for a 2-reactor system, with a capacity of 41,600 tonnes/year digestate fibre, representing 68% of the total produced in the county. This results in a digestate fibre processing cost of £35.70/tonne. Due to the higher production of digestate fibre and larger area of Lincolnshire, the inclusion of transportation costs had a more significant effect.

Figure 6.3 shows that when transportation costs were included, increasing the HTC plant size beyond 3 HTC reactors began to increase the digestate processing cost, but the cost differential was relatively small comparing 2, 3 and 4 reactors. Beyond 4 reactors, the processing costs including transportation started to rise more significantly. A 3-reactor system would process 62,400 tonnes of digestate fibre a year, 61% of the total produced in Lincolnshire. The minimum cost came to £37.10/tonne of digestate fibre processed.

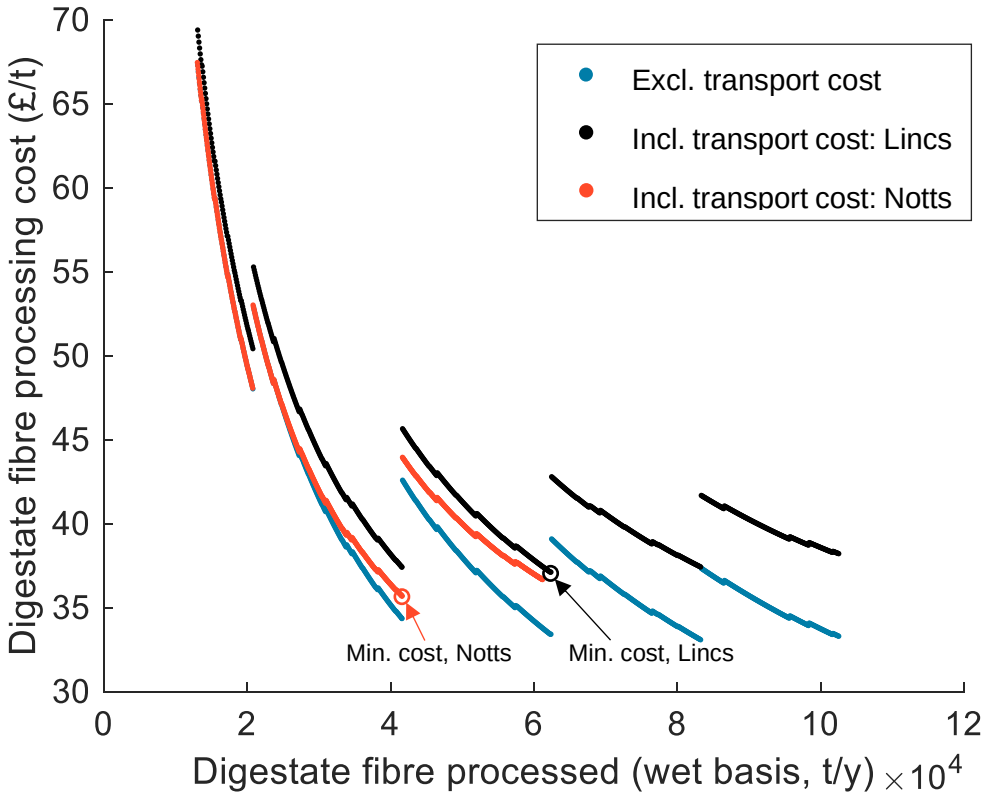


Figure 6.3: Effect of excluding or including digestate fibre processing cost using Lincolnshire and Nottinghamshire as example counties. Each step change comes from an increase in the number of reactors in the plant.

Looking ahead, the optimum locations for HTC plants found for Nottinghamshire and Lincolnshire represent promising locations for the future of AD production. They are both close to the largest cities in the counties (Nottingham and Lincoln) and therefore any increase in food waste AD in the country is likely to be situated nearby. The trade-off between plant capacity and location here adds depth to the analysis performed in Section 5.7 Plant Capacity in Chapter 5 HTC Plant Model and highlights the kinds of considerations that should be made when assessing the feasibility of waste treatments. The economy of scale and generation of waste must both be taken into account in order to develop the lowest cost solutions.

6.2.2 Digestate dewatering

As discussed in Section 5.6 Optimisation from Chapter 5 HTC Plant Model, processing dewatered digestate fibre is much cheaper than whole digestate in terms of the CAPEX and OPEX of the HTC plant. Because of these large cost savings, dewatering of whole digestate is almost considered an essential precursor to HTC and therefore the cost and logistics are important to consider. Digestate fibre typically has a moisture content of 75 wt.% and estimates for nutrient content were taken from a report by the Federal Ministry for Sustainability and Tourism (BMNT) in Austria [22]. Solid recovery and cost estimates were taken from a report from the Waste and Resources Action Programme (WRAP) in Wales [20]. Details of these are given in Section 3.4.2 Feedstock Input and Dewatering in Chapter 3 Methodology.

Publicly available data on digestate dewatering is sparse but WRAP sought to investigate the dewatering of food waste-based digestate in their study [20]. Mechanical dewatering of sewage sludge in the waste water treatment industry is commonly used, but dewatering food waste-based digestate presents technical and economic challenges due to its surface properties [192]. The WRAP study conclusions indicated a heavy dependence on the level of dilution of the food waste prior to being processed within the AD plant. They identified this as a critical assumption and so used two central cases in their analysis, one where the food waste was diluted to 20 wt.% dry solids (central case 1) and another where it was diluted to 10 wt.% dry solids (central case 2). The resulting whole digestates, before dewatering, had 5.6 wt.% dry solids and 2.8 wt.% dry solids respectively. For central case 1, none of the 7 dewatering scenarios offered cost benefits compared to the baseline: the direct application of whole digestate to land. However, for central case 2, the majority of dewatering scenarios offered savings compared to the baseline. The use of a centrifuge and biological oxidation of the digestate liquor followed by disposal to the watercourse was the lowest-cost solution.

The costs in the WRAP report were reported per quantity of feedstock sent to AD, which in their case was food waste with a dry solids content of 26 wt.%. The WRAP report calculated synthetic fertiliser displacement savings for each scenario. That is the expected financial saving made from spreading digestate to land and a reduction in synthetic fertiliser purchase, based on the nutrient content of the digestate. However, the report uses food waste digestate only and assumes that spreading to land is and will remain unrestricted. As discussed earlier, the nutrient content varies widely depending on the feedstock used and there are concerns about the direct application of whole digestate and/or digestate fibre to land. Therefore, for the purpose of this work, the costs were calculated both including and excluding these fertiliser replacement savings.

Table 6.2: Cost comparison of the baseline case and lowest-cost dewatering scenario for both central case 1 and central case 2 as reported by WRAP [20] *using a 2-reactor HTC system processing 41,600 tonnes of digestate fibre annually as a basis.*

	Central case 1 (digestate 5.6 wt.% dry solids) [20]		**Central case 2 (digestate 2.8 wt.% dry solids)** [20]		**BMNT** [22]
	Baseline	Dewatering	Baseline	Dewatering	
Whole digestate (t/y)	218,300		436,700		218,300
Digestate fibre (t/y)		41,600		41,600	41,600
Digestate liquor (t/y)		176,700		395,100	176,700
Whole digestate spread to land (GBP/y)	£2,934,000		£5,804,000		
Centrifuge (GBP/y)		£1,066,000		£2,131,000	£197,000
Biological oxidation + disposal to water course (GBP/y)		£871,000		£1,947,000	
Digestate fibre spread to land (GBP/y)		£617,000		£617,000	
Total (excl. synthetic fertiliser replacement) (GBP/y)	£2,934,000	£2,554,000	£5,804,000	£4,696,000	
Total (incl. synthetic fertiliser replacement) (GBP/y)	£1,940,000	£2,341,000	£4,789,000	£4,483,000	

Table 6.2 shows a cost comparison for dewatering for both central case 1 and central 2, including and excluding the savings associated with synthetic fertiliser replacement. Centrifuge capital costs were converted to a yearly amount using a 20-year lifespan. The costs have been calculated where digestate fibre production would amount to 41,600 tonnes per year, the throughput of a 2-reactor HTC plant. These results highlight the importance of the dilution assumption and also whether the application of whole digestate or digestate fibre to land offsets synthetic fertiliser costs. The results also show that dewatering is a relatively expensive process that cannot necessarily be omitted when assessing the economic feasibility of the HTC of digestate. The clearest comparison that can be made is the calculated cost of spreading digestate to land (excluding synthetic fertiliser replacement) at £617,000 for 41,600 tonnes of digestate fibre, equating to a cost of £14.83/tonne of digestate fibre. This is in line with the £16/tonne estimate made earlier using the WRAP report, which assumes that regulations associated with the spreading of digestate to land are unchanged. If further restrictions were to be introduced, as is expected over the coming years, the cost of spreading

could increase further by requiring extra processing or an outright ban on its spreading and so a costly hazardous waste disposal cost.

There are further observations to make about the dewatering cost calculations, indicating that they give a pessimistic view. As food waste digestate is more challenging to dewater than other typical sludges [192], dewatering costs would be lower for the waste-based and farm-based digestates considered. As a comparison, the centrifuge CAPEX and OPEX was recalculated using the values reported by the BMNT government body in Austria, based on the dewatering of household and green waste [22]. While these were cruder than the figures reported by WRAP, the yearly cost (assuming a CAPEX split over 20 years) came to just £197,000, a fraction of the £1,066,000 estimated using the WRAP results. WRAP gave a number of reasons why the dewatering of food waste digestate is more challenging and therefore more costly. The dewatering of sewage sludge digestate and farm manures is often performed using cheaper technologies such as screw presses and belt filters. However, testing has shown that the filtering of food waste digestate is particularly difficult, reducing its suitability for these types of cheaper technologies. Additionally, the digestate liquor separated from food waste during the dewatering process is likely to contain higher levels of organic and inorganic material than from sewage sludge digestate. This means that the cost estimation of biological oxidation and disposal to watercourse is at the higher end of cost estimates and is expected to be lower for other digestate feedstocks.

Ultimately, from the sizing and costing of the HTC plant, detailed in Chapter 5 HTC Plant Model, Section 5.6 Optimisation, the dewatering of digestate is highly recommended. However, it represents a significant cost and so should be considered to understand the true economic feasibility of the HTC of digestate. There is a paucity of information on the cost of dewatering, meaning that formulating an accurate estimate is difficult. Using the WRAP report gives a total annual dewatering cost of over £2 million, a notable amount when compared to the £1.2 million OPEX of an HTC plant treating the resulting digestate fibre. However, this WRAP estimate is based on technologies from 2014. Dewatering technologies for digestate are still developing and so the price is expected to fall over the coming years. Without more robust cost data on the dewatering of all types of digestate, it is difficult to calculate better estimates than done so in this work. For HTC to be profitable, it should compete with the cost of spreading to land, which comes in at £14.83 in this report. With an expected increase in this cost, HTC may prove as an interesting alternative treatment.

6.3 Market analysis

As discussed in Chapter 3 Methodology Section 3.7 Market Analysis, there are two main revenue streams for the HTC of waste biomasses: the sale of HTC products and savings in

feedstock treatment compared to current management and/or disposal techniques. The sale prices of the different HTC products in Table 3.8 in Chapter 3 Methodology have been adapted for a comparison between the different types of digestate, displayed in Table 6.3. The carbon content and thus energy content of crop-based and farm-based hydrochars were much higher than that of the waste-based digestate. Therefore, if hydrochar is sold as a solid fuel on an energy basis, its sale price is much higher. Additionally, if the hydrochar is to be used as a form of soil amendment for carbon sequestration, the crop-based treated hydrochar would have a higher sale price due to its greater carbon content. It is important to note that the yield of hydrochar and treated hydrochar was found to be higher for the crop-based and farm-based digestates.

Table 6.3: Equivalent sale prices and comparative costs for digestate and a range of HTC products for each of the digestate types.

Product market		Sale price	Equivalent hydrochar sale price (£/tonne)			Ref.
			Waste-based	Farm-based	Crop-based	
Digestate, saving			£16/tonne	£16/tonne	£16/tonne	[21]
Hydrochar, solid fuel	Mass basis	£150/tonne average	£150/tonne	£150/tonne	£150/tonne	[136]
	Energy basis	£8.46/GJ	£99/tonne	£177/tonne	£181/tonne	
Hydrochar, fertiliser		£0.61/kg N £1.44/kg P £0.56/kg K	£64/tonne	£79/tonne	-	[167]
Treated hydrochar, carbon sequestration		£418/tonne C	£89/tonne	£134/tonne	£147/tonne	[168]
Process liquid, fertiliser		£0.61/kg N £1.44/kg P £0.56/kg K	£5/tonne	£6/tonne	-	[167]

The applicability of hydrochar products in each of these target markets have been discussed in Chapter 4 HTC Test Programme, Section 4.3.4 Product use suitability. In summary, hydrochar as a solid fuel should be low in ash to prevent issues with slagging and fouling. Hydrochar applied to land for either carbon sequestration or as a fertiliser should meet the limits set out in PAS110 [10], not exceed nitrogen limits within Nitrate Vulnerable Zones (NVZs) [193], and meet the biochar standards set out in the European Biochar Certificate [92] and Biochar Quality Mandate [170].

6.4 Investment analysis

An investment analysis was performed for a HTC plant treating 41,600 tonnes of digestate fibre using the transportation costs associated with Nottinghamshire as default. Constants used in this analysis were defined as shown in Table 6.4.

Table 6.4: Constants defined for the investment analysis.

	Value
Plant life	20 years
Discounted rate of return	10%, the lower end of the range of hurdle rates for biomass conversion technologies as HTC is still an emerging technology [194]
Corporation tax	19%, UK value
Annual inflation	2%
Linear depreciation period	10 years
Production start	2nd year of operation
Default HTC plant conditions	Temperature: 210 °C Residence time: 180 minutes Feedstock moisture content: 75 wt.% Plant capacity: 41,600 tonnes (wet) Feedstock transportation model: Nottinghamshire

An investment analysis was performed for the three types of digestate tested experimentally. The default prices from Table 6.3 were used. This scenario had a hydrochar sold as a solid fuel and disposal of the process liquid.

The cumulative net present value over the period of 20 years is shown in Figure 6.4. The first two years represent the capital investment into the HTC plant with operation beginning in the 2nd year. This shows that with the default conditions, the HTC of waste-based digestate did not break-even in the first 20 years. The farm-based digestate and crop-based digestate however performed better, albeit with payback periods in excess of 10 years. A higher yield of hydrochar increased the income of the HTC plant due to increased sales, with the crop-based digestate resulting in the largest revenue. As the farm-based and crop-based hydrochars had higher energy contents, their sale price was higher on an energy-basis; the values are given in Table 6.3. The dashed lines in Figure 6.4 show how the economic performance for the farm-based and crop-based digestates improved, but the waste-based digestate was less profitable. Without a clear definition of how the characteristics of hydrochar affect its sale price, this is something to consider.

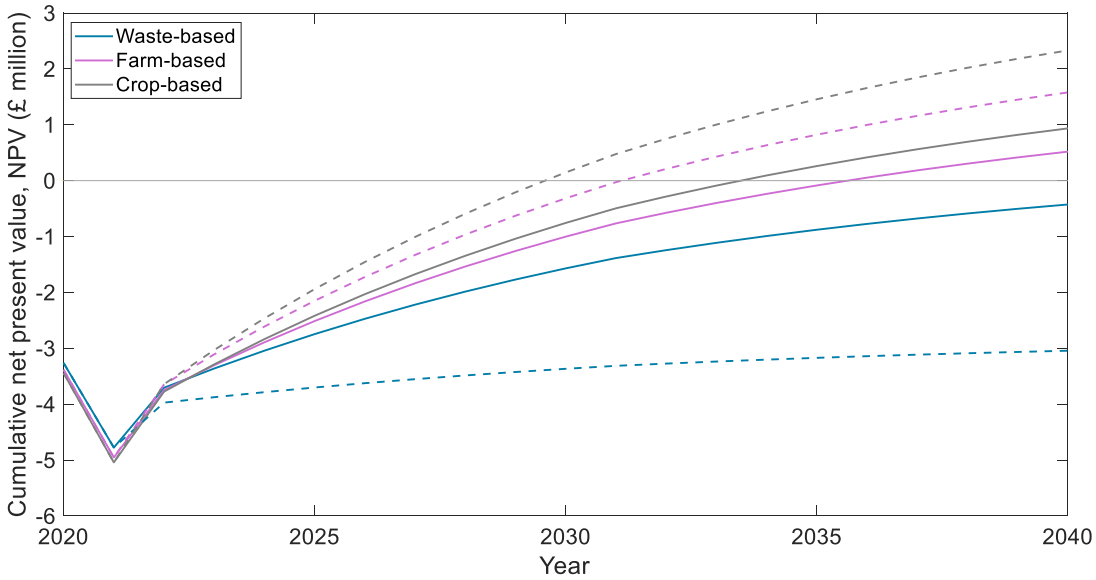


Figure 6.4: Cumulative net present value for the three digestates over 20 years: waste-based, farm-based and crop-based. The default plant conditions (Table 6.4) were used. The solid line represents hydrochar sold as a solid fuel at £150/tonne. The dashed line represents hydrochar sold on an energy content basis (£99/tonne, £177/tonne and £181/tonne, respectively, Table 6.3). The process liquid was disposed of. The digestate saving was set to £16/tonne.

The other product markets were also investigated (Figure 6.5). The same default conditions as before were used, with the waste-based digestate as an example. This highlights that selling hydrochar as a fuel was the best option. Extra revenue could be generated through the sale of process liquid as a fertiliser product. Having these two revenue streams resulted in a payback period of just under 15 years. The least economically favourable option was using the hydrochar as a fertiliser. Hydrochar as a carbon sequestration material did not perform well either. This is partially due to the torrefaction unit required to produce hydrochar suitable for soil application, representing an increase in CAPEX.

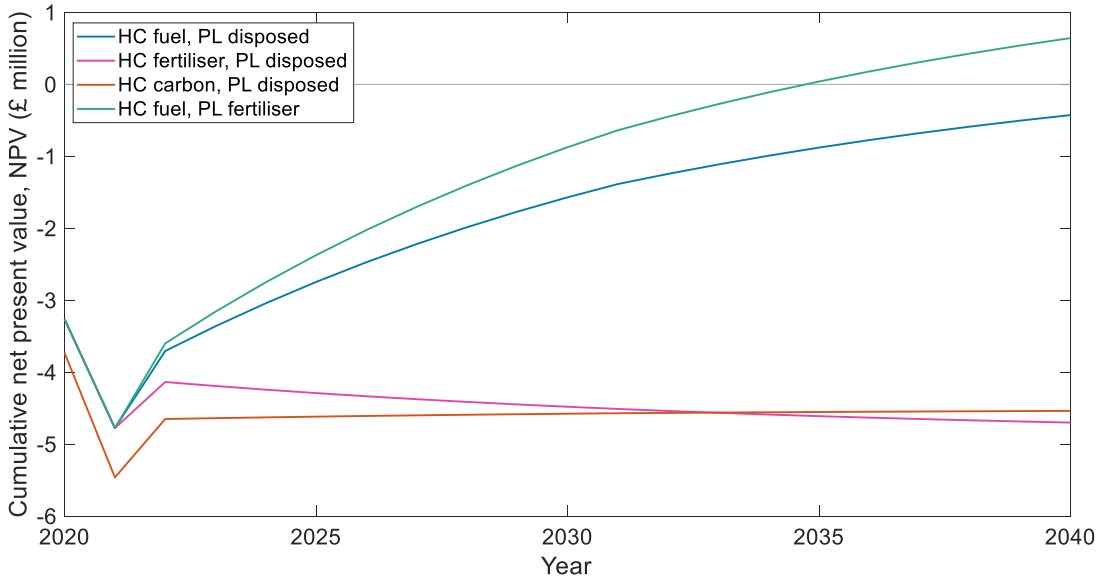


Figure 6.5: Cumulative net present value for the waste-based digestate over 20 years. The default plant conditions (Table 6.4) were used. Four different market conditions were investigated: hydrochar sold as a solid fuel, a fertiliser product, or for carbon sequestration, and the process liquid either disposed of or used as a fertiliser. Prices are shown in Table 6.3. The digestate saving was set to £16/tonne.

6.4.1 Sensitivity analysis

The payback period was used as a tool for measuring the economic feasibility of the process. While using the process liquid as a fertiliser rather disposing of it through waste water treatment did increase the economic performance, the viability of HTC mostly depends on two variables: the sale price of hydrochar and the cost savings of the feedstock. As neither of these markets are fully established, their effect is key to understanding the HTC of digestate. In order to investigate this, the payback period was fixed, and the required feedstock cost and hydrochar sale price were calculated. A negative feedstock costs represents a saving.

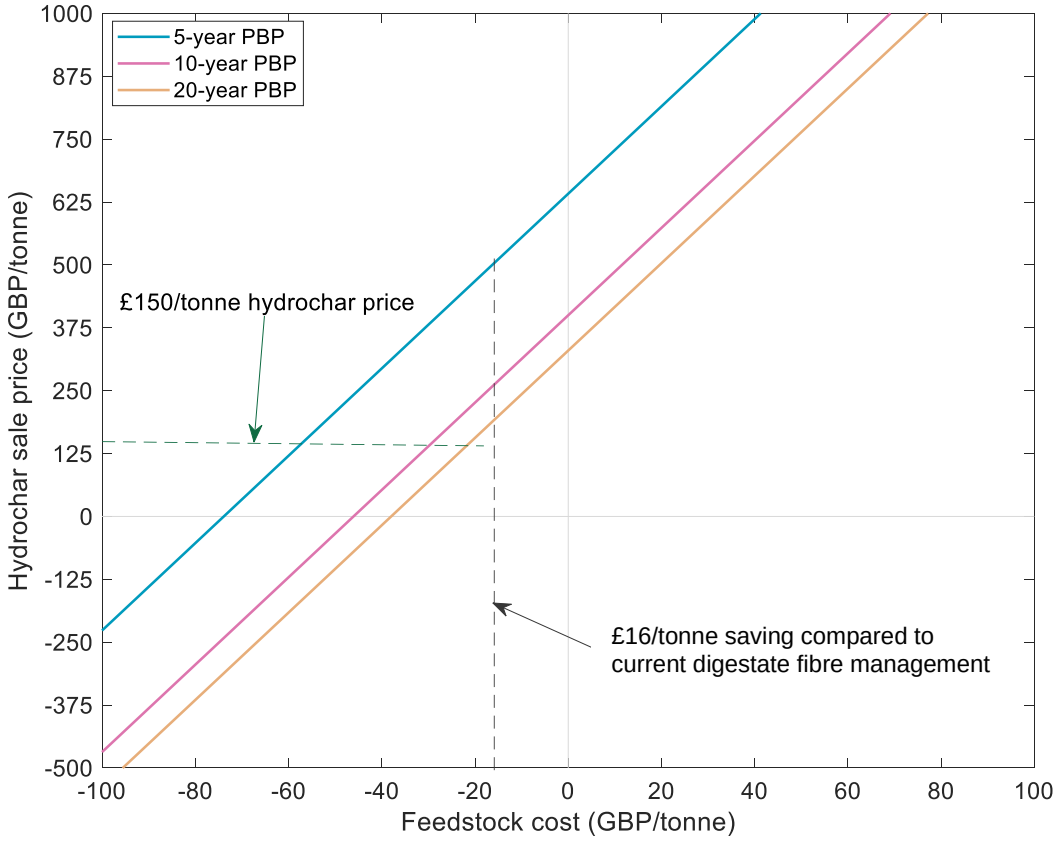


Figure 6.6: Required feedstock costs and hydrochar sale price to achieve payback periods of 5, 10 or 20 years for the waste-based digestate. The default plant conditions (Table 6.4) were used. A negative feedstock cost represents the saving available by diverting the feedstock from its current management/disposal method.

Figure 6.6 shows the costs required for an HTC plant processing the waste-based digestate to achieve a payback period of 5, 10 or 20 years. As expected, for a shorter period payback period, a higher hydrochar sale price and/or a lower feedstock cost were required. As shown in Figure 6.6, the current plant with a hydrochar sale price of £150/tonne and a digestate saving of £16/tonne did not result in a viable process. These were used as benchmarks to see what value of digestate saving or hydrochar sale price was required for a defined payback period. The results for the waste-based digestate are shown in Table 6.5.

Table 6.5: Required digestate savings and hydrochar sale prices when fixing one value to achieve payback periods of 5, 10 or 20 years for the waste-based digestate. It is assumed that process liquid is disposed of and not sold as a fertiliser.

Payback period	Required digestate fibre saving (£/tonne) (Hydrochar sale price fixed at £150/tonne)	Required hydrochar sale price (£/tonne) (Digestate savings fixed at £16/tonne)
5 Year	58	500
10 Year	30	255
20 Year	21	188

The payback period was assessed for the other types of digestate too. As seen in the investment analysis (Figure 6.7), the waste-based digestate was the worst performing and higher hydrochar sale prices would be required to achieve a payback period of 10 years. However, due to the lower quality and higher risk of contamination for waste-based digestate, its management and disposal costs are expected to be greater than that of the farm-based and crop-based digestate, offering greater savings for the HTC operator.

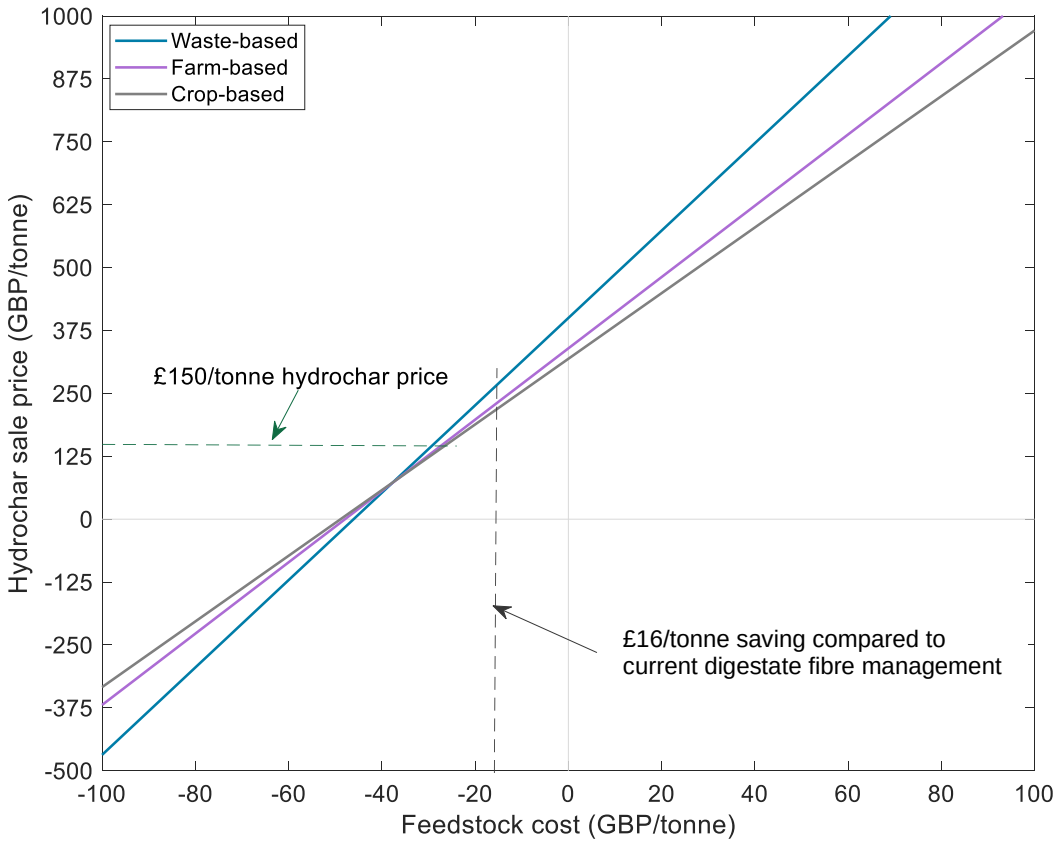


Figure 6.7: Required feedstock cost and hydrochar sale price to achieve a payback period of 10 years. The default plant conditions (Table 6.4) were used. A negative feedstock cost represents the saving available by diverting the feedstock from its current management/disposal method.

While the CAPEX was validated with industrial data, the estimate was made using the factorial method detailed in Seider et al. with an accuracy of ±30% [154]. A reduction or increase in

CAPEX only led to a difference in the initial investment in the first two years, seen in Figure 6.8. The system where the CAPEX was reduced by 30% resulted in a payback period of fewer than 14 years.

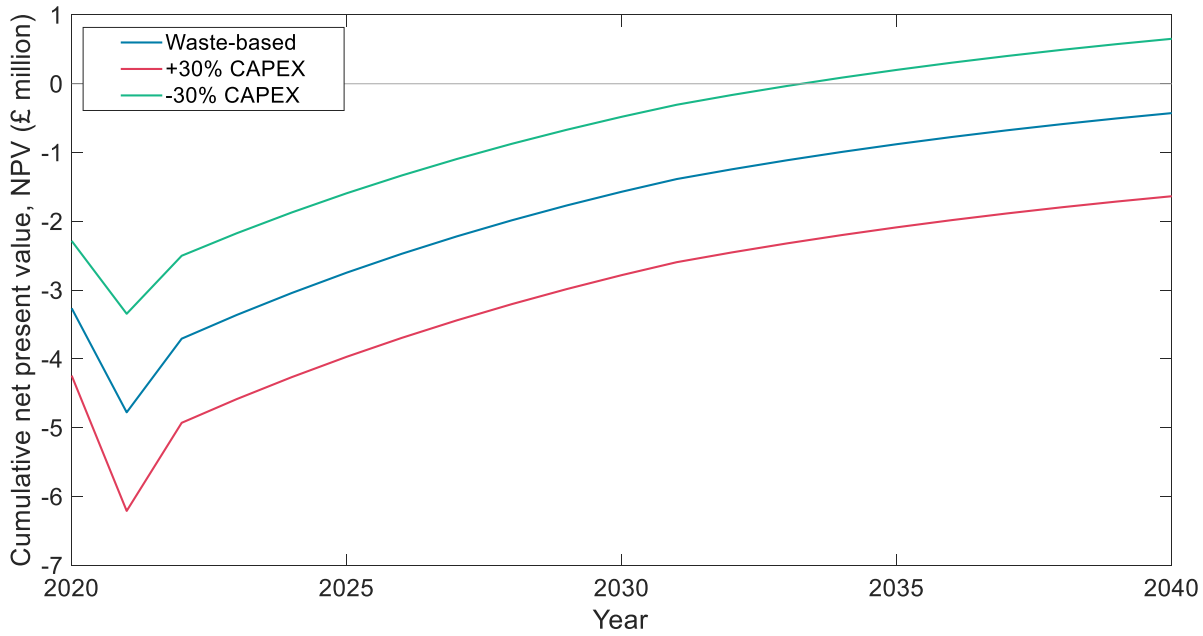


Figure 6.8: Cumulative net present value for the waste-based digestate over 20 years. The default plant conditions (Table 6.4) were used. Hydrochar was sold as a solid fuel for £150/tonne and process liquid was disposed of. The digestate saving was set to £16/tonne. The effect of CAPEX was investigated using the ±30% included in the estimate.

6.4.2 Alternative feedstocks

As demonstrated in Chapter 5 HTC Plant Model, the process model was capable of assessing the feasibility of other feedstocks in addition to the digestates. Miscanthus and wood pellets were selected as typical lignocellulosic feedstocks for comparison with the digestate feedstocks. From the results of experimentation and regression models, these feedstocks resulted in higher yields of hydrochar with a higher energy content. Therefore, the revenue from hydrochar sales would be higher. However, miscanthus and wood pellets are both higher-value feedstocks in themselves, and rather than offering a saving, the operator would have to pay for their use. The wood pellets feedstock cost was set as £80/tonne [195] and the miscanthus feedstock cost was set at £50/tonne [196]. The effect of a feedstock cost rather than saving is displayed in Figure 6.9. Here, it is clear that with the current sale price of hydrochar, the HTC of miscanthus or wood pellets is not economically favourable, even with a larger yield of hydrochar. Even when the hydrochar is priced on an energy-basis, shown by the dashed lines, the HTC of miscanthus and wood pellets did not result in a viable economic model.

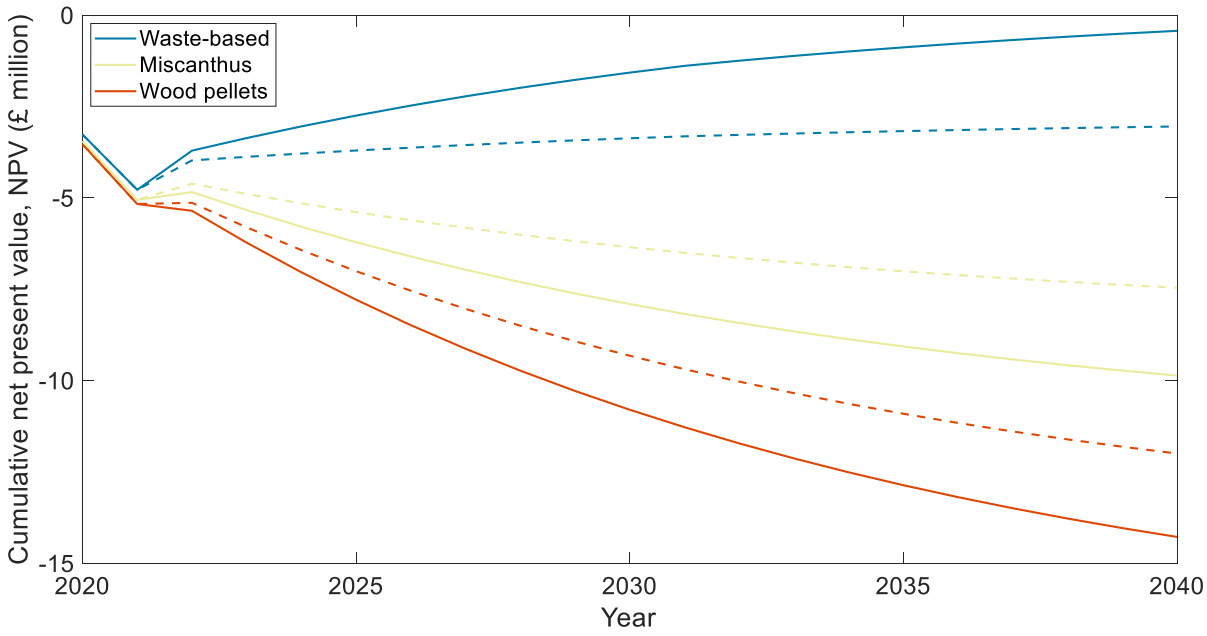


Figure 6.9: Cumulative net present value for waste-based digestate, miscanthus and wood pellets over 20 years. The default plant conditions (Table 6.4) were used. The solid line represents hydrochar sold at £150/tonne. The dashed line represents hydrochar sold on an energy content basis (Table 6.3). The process liquid was disposed of. The digestate saving was set to £16/tonne, miscanthus cost as £50/tonne and wood pellets cost at £80/tonne.

6.5 Conclusions

The AD market in the UK is growing and it is predicted that the AD of food waste in particular will increase in the UK over the next 10 years. While many farm-based AD operators spread digestate on their own land, commercial and industrial AD operators often have to pay for its removal and management. The current cost for the transportation and land application of dewatered digestate fibre with a 25 wt.% solids content comes to £16/tonne. This cost is expected to rise with the introduction of tighter restrictions on the application of digestate to land and stricter storage requirements. Therefore, there is demand for an alternative, low-cost treatment for digestate in the UK, with HTC as a potential candidate. The economic study presented here is the first time the viability of HTC as a digestate treatment in the UK has been assessed. It is an improvement over the existing publicly available HTC economic models as it was designed as fully flexible and validated with industrial data.

Due to the high moisture content of digestate, an important consideration is the cost of its transportation to the HTC site. Two counties were selected as examples: Lincolnshire, the largest producer of digestate, and Nottinghamshire, the county with the highest density of digestate production to area. The optimum location of HTC plants in these counties was modelled and the cost of transporting digestate to this site was calculated. Both counties showed similar relationships, with a slightly lower transportation cost found in Nottinghamshire. Despite savings due to economies of scale, it was found that a 2-reactor

HTC plant capable of treating 41,600 tonnes of digestate fibre a year gave the lowest costs on a unit basis. This plant size was taken forward for the rest of the analysis.

As found in Section 5.6 Optimisation from Chapter 5 HTC Plant Model, treating dewatered digestate fibre as opposed to whole digestate offered large savings in CAPEX and OPEX of the HTC plant. The mass balance and nutrient content resulting from dewatering was included in the process model and detailed in Section 3.4.2 Feedstock Input and Dewatering in Chapter 3 Methodology. This chapter assesses the economics of dewatering. Using the results from the WRAP report [20], dewatering enough whole digestate to produce 41,600 tonnes of digestate fibre would cost in the region of £2–5 million, depending on the assumptions used. They reported the cost of spreading the digestate fibre to land as £14.83/tonne, within the same range of the £16/tonne previously calculated. However, there is very limited information on the cost of digestate dewatering and the WRAP report was focussed on the dewatering of food waste digestate, known to be more complex and costly than other forms. As it is difficult to gain an accurate estimate of the cost of dewatering different types of digestate, further calculations are recommended on a specific case basis.

A series of investment analyses were performed to understand the economic feasibility of using HTC for the treatment of digestate. The majority of industrially operating HTC processes are based on specific arrangements and contracts with waste producers. The purpose of this work was to take a more comprehensive approach to understand the long-term viability of the technology. This work generally considered pessimistic scenarios so as to not overestimate the benefits of HTC. As it stands, with available savings of £16/tonne of digestate fibre and a hydrochar sale price of £150/tonne, a HTC plant treating 41,600 tonnes of digestate fibre annually offers a payback period of roughly 14 years for farm-based and crop-based digestate (Figure 6.4). For waste-based digestate, the payback period is greater than 20 years. These differences are due to the increased yield of hydrochar from farm-based and crop-based digestate. However, the calculations did not account for the difference in product quality between the feedstocks. Details on the suitability of HTC products for different markets are given in Chapter 4 HTC Test Programme. The hydrochar from farm-based and crop-based digestate had a noticeably higher energy content and therefore a higher sale price if sold on an energy basis (Table 6.3). It is difficult to predict with certainty which type of digestate for HTC would be the most profitable. Waste digestate contains more contaminants than other forms of digestate and therefore is likely to have more stringent restrictions on its spreading to land, incurring any increase in management and disposal costs. It could offer larger savings compared to farm-based and crop-based digestate. Waste-based digestate is also more likely to result from commercial and industrial AD operators who do not have the option of spreading to their own land, unlike small-scale farm-based plants. This means that the bulk of large-scale

plants must pay for the removal of digestate generated at their plant and so will be more interested in alternative treatments. With a hydrochar sale price fixed at £150/tonne, waste-based digestate would need to offer savings of £61/tonne or £30/tonne for a 5- or 10-year payback period, respectively. As farm-based and crop-based digestates yielded higher quantities of hydrochar, their savings would only need to be £27/tonne for a 10-year payback period. The difference grows when the product qualities from the different digestates are accounted for (Table 6.3). Farm-based and crop-based digestates result in higher hydrochar sale prices, both as a fuel and also as a means for carbon sequestration. However, marketing hydrochar for carbon sequestration resulted in lower sale prices, with a fixed price of £418/tonne of carbon. This was true for all feedstocks and was observed for the waste-based digestate in Figure 6.5. Selling hydrochar as a fertiliser was the worst scenario, creating annual losses for the HTC plant. Selling hydrochar as a solid fuel and the process liquid as a fertiliser was the most promising model, although this would only be applicable to the crop-based digestate as the other two are not considered suitable as solid fuel products.

An investment analysis of the HTC of lignocellulosic feedstocks highlighted the importance of feedstock cost or savings on the economic feasibility of the process. The HTC of both miscanthus and wood pellets led to annual losses, even when the higher hydrochar energy content and therefore higher potential sale price was taken into account. Feedstock cost has been highlighted as the most influential factor for the profitability of HTC [133,134], as discussed in Chapter 2 Literature Review. This is reflected in this work too, through the analysis of payback periods and the investment analysis for alternative feedstocks. At a digestate saving of £16/tonne, HTC is not an economically favourable treatment for waste-based digestate and results in large payback periods for farm-based and crop-based digestates. However, with an expected increase in digestate management costs, HTC could present itself as a promising alternative. As HTC is more suited to treating digestate fibre, with a moisture content of 75 wt.%, as opposed to whole digestate, typically with a moisture content of 97–94 wt.%, the cost of dewatering would have to be considered by any operator. If the restrictions were tightened to the point where land application of digestate was prohibited, meaning it was sent to landfill at a cost of £113/tonne, HTC would certainly offer cost savings over digestate disposal.

In summary, the profitability of the HTC of digestate hinges on a number of key factors, and in the current market can achieve payback periods of fewer than 15 years under certain circumstances. Looking ahead, its economic potential is likely to grow and HTC will become an increasingly attractive alternative technology to digestate spreading to land. Understanding this is important when considering the level of support to be given to HTC for digestate

treatment in the UK and the principles and methodology covered in this study can be applied further afield.

7 Conclusions and Future Work

7.1 Introduction

This work has been focussed on testing, modelling and assessing HTC and its potential for wet biomass treatment at an industrial scale, making important contributions to the HTC research and development field. Experimental work was carried out on a range of the most common types of digestate generated in the UK. Regression models were determined, describing the relationship between HTC product properties and HTC conditions. These regression models formed part of a comprehensive process model made up of a range of unit operations, based on industrial-scale HTC plant plans. The process model developed in this work was validated with industrially sourced data and has been shown to perform better than currently available literature models. An interactive Matlab App incorporating the process model was created, allowing for the user to alter HTC operating and/or market conditions and assess the impact on HTC products and economic performance. This fully flexible and functional app is the first of its kind in the field and significantly improves the ease at which one can assess a range of HTC plant options. The process model was used as the basis of a more in-depth study, considering the role HTC of digestate could play in the UK. This is a timely analysis as the prevalence of digestate generation in the UK increases, yet further regulations and restrictions on its proper management and disposal are expected. Overall, this work presents an improvement of the current understanding of the HTC process and economics.

This chapter concludes each of the areas studied, summarising the current understanding, the gaps in research, and the key results. Additionally, suggestions for future work have been made, useful for a more in-depth and targeted study of HTC for a particular feedstock or market.

7.2 Current status of hydrothermal carbonisation

HTC can be dated back to 1913, however it has only been in the past 15 years that research and commercial interest has really taken off. The first HTC patents were granted in 2008, and there are currently a number of industrial-scale HTC plants based in Europe and Asia [27–29,197]. These typically operate with specific contracts with local authorities or businesses for the treatment of a particular waste. Therefore, more still needs to be understood of the general feasibility of HTC under a range of conditions and markets.

Both feedstock type and HTC operating conditions affect the product properties. Most research has focussed on the HTC temperature and residence time as the two main variables. Generally, results show that an increase in temperature and/or time leads to an increase in

the degree of carbonisation of hydrochar, that is a higher carbon and energy content, but a lower mass yield. Of the two, temperature has been shown to be the most influential factor [42,44,45,47,50,52]. Other commonly researched process conditions include pH and initial solids content of the feedstock. Both an increase in pH and initial solids content have been found to increase the degree of carbonisation [56,57], but some studies have concluded that the initial solids content is insignificant [44,47,50]. Understanding the HTC reaction and pathways can lead to better informed choices on the process conditions, but ultimately industrial decisions are most often based on working limits and operating costs as well as the trade-off between higher hydrochar yield and higher quality hydrochar. Hydrochar markets are still developing, and the optimisation of product properties still very much depends on the market of interest. The process liquid is also increasingly considered as a valuable product and so more needs to be understood about its properties too. This highlights the need for a clear understanding of the techno-economics of HTC, rather than just the details of the reaction pathways and products. It is believed that knowing more about the economic potential of HTC will aid its commercial development.

7.3 Modelling HTC

The HTC process and techno-economics have been modelled using various approaches. Most commonly seen in literature are statistical models based on the process severity, that is the HTC temperature and time. Multiple linear regression models have been developed to describe the dependence of product yields and composition on the process conditions for a particular feedstock. Some researchers have gone on to develop universal models, applicable for all types of feedstocks but these have limited accuracy. As well as these HTC process models, there are a number of techno-economic models for HTC. However, when compared with industrial data, there appears to be a number of large discrepancies.

Based upon these observations, this work involved the creation of a regression model for specific feedstocks, to ensure accuracy, followed by the development of a techno-economic model validated by industrial data. HTC was performed on three types of digestate feedstock sourced in the UK: waste-based, farm-based and crop-based. These samples were deemed representative of the most common types of digestate in the country.

7.3.1 Regression model

Temperature, residence time and feedstock moisture content were assessed. Chapter 4 HTC Test Programme details these experimental and regression model results. For the waste-based digestate, a Box-Behnken design of experiments was used, requiring 15 HTC runs at different conditions. Fewer HTC runs were performed for the farm-based and crop-based

digestates over the full range of conditions. The regression models created from the experimental results agreed with literature in that HTC temperature had the largest effect, with an increase in temperature leading to a decrease in hydrochar yield but an increase in its carbon content. The waste-based, farm-based and crop-based digestates had carbon contents of 27.6 wt.%, 38.6 wt.% and 39.9 wt.% on a dry basis and 43.5 wt.%, 50.1 wt.% and 44.0 wt.% on a dry ash-free basis, respectively. The biggest difference between the samples was the ash content, with the waste-based digestate containing 36.5 wt.%, compared to 22.9 wt.% for the farm-based digestate and only 9.3 wt.% for the crop-based sample. The yield of hydrochar from waste-based digestate was the lowest, followed by the farm-based and then crop-based digestates. The maximum carbon contents of the hydrochars were 29.5 wt.%, 49.5 wt.% and 59.6 wt.% on a dry basis and 58.6 wt.%, 73.8 wt.% and 67.8 wt.% on a dry ash-free basis for the waste-based, farm-based and crop-based digestates, respectively. As expected, the regression models developed here gave much more accurate predictions for the feedstocks used experimentally than the generic models found in literature. Generally, the regression models for the waste-based digestate gave a better fit than for the other types of digestate, as expected from the larger number of experimental runs. However, the goodness of fit for the farm-based and crop-based digestate models were deemed satisfactory to be used in the process model.

The effect of three factors temperature, residence time and feedstock moisture content were investigated experimentally. This methodology followed what is commonly seen in literature and meant that the HTC process was better understood over a range of process conditions. During the course of this work, insights were gained from a number of industrial HTC operators. It became clear that at industrial scale, residence time is often determined by plant equipment geometry and feedstock moisture content is limited to <75 wt.% as to avoid the need for overly large plant equipment processing very wet waste, coming at a high capital cost. Therefore, it is suggested that if an industrially based process model is desired, the experimental design could be improved to focus on the effect of temperature alone with a fixed, or limited range, of residence time and moisture content. This could reduce the number of experimental runs performed but would limit the understanding of the effect of multiple process conditions on HTC products.

The carbon, hydrochar and nitrogen contents of the hydrochars and process liquids were measured using a CHN analyser and the oxygen was calculated by difference. The concentrations of inorganic elements were measured using ICP analysis. The measured recovery of carbon, phosphorus and potassium in the HTC products generally ranged between 90% and 110%, suggesting a reasonably good estimate. However, the measured recovery of nitrogen in the hydrochar and process liquid totalled between 60% and 140%. This apparent

error could simply be due to nitrogen loss during the preparation of samples and highlights the need for repeated, more robust testing in order to gain an accurate estimate of elemental concentrations.

7.3.2 HTC product use

The most established market for hydrochar is solid fuel. But, from the three types of digestate studied, it was deduced that only the crop-based digestate showed true potential for the production of a solid fuel due to its low ash content. The measured ash content of hydrochar from waste-based and crop-based digestates exceeded 30 wt.%. Such a high ash content would likely cause multiple issues, such as slagging and fouling, in a standard pulverised fuel boiler.

Alternative hydrochar markets include fertiliser and carbon sequestration. The nutrient contents of the hydrochars produced from the waste-based and farm-based digestates were analysed. Compared to the digestate feedstocks, the hydrochars contained a higher concentration of phosphorus and a similar concentration of nitrogen, although most of the potassium was found in the process liquid. Additionally, the concentrations of trace elements generally did not exceed biochar standards. Literature has shown that raw hydrochar has phytotoxic properties, but that post-treatments can alleviate these. To explore this, a large quantity of hydrochar from a demonstration-scale HTC plant was sourced. It was produced from a waste-based digestate independent of that used in the HTC runs. The hydrochar was subject to thermal post-treatment at 275 °C and used in germination and plant trials. It was found that there was still significant plant growth, even with loadings of treated hydrochar of up to 30 vol.%. While further study is required, this suggests that the application of hydrochar to land is a viable option and is unlikely to result in complete germination inhibition. This could be instrumental in exploring future options for carbon sequestration in land.

The nutrient content of the process liquids from the waste-based and farm-based digestates was also measured. Again, these were found in measurable quantities, opening up the fertiliser market. The process liquid from the farm-based digestate had the highest nutrient content.

7.3.3 Techno-economic model

A process model was created in MATLAB to represent an industrial-scale facility, with details included in Chapter 5 HTC Plant Model. The model consisted of a series of unit operations covering all stages of the HTC process. It was designed with flexibility in mind, with user-defined feedstocks, process conditions, plant size, location, markets and product sale prices.

The default plant conditions were defined as 215 °C for a residence time of 180 minutes, treating 41,600 tonnes of feedstock a year with a moisture content of 75 wt.%. These were selected to represent typical industrial conditions for a 2-reactor HTC plant.

Using the regression models developed in earlier work, the default plant was predicted to produce 4,800, 5,900 or 6,300 tonnes of dry hydrochar per year for the waste-based, farm-based and crop-based digestates, respectively. Engineering equations were used to calculate the expected energy use under these conditions, coming to 800–1,000 MWh/year of electrical energy and 8,100–10,000 of thermal energy MWh/year. Publicly available cost data was used to estimate the capital expenditure (CAPEX) and operating expenditure (OPEX) of the plant. The CAPEX fell within the range of £5.5–5.8 million for each of the feedstocks. The size of the hydrochar dryer and thermal post-treatment equipment had to be larger to accommodate the larger volume of hydrochar produced from the farm-based and crop-based digestates, explaining the higher CAPEX for these feedstocks. The CAPEX included £675,000 for an optional thermal post-treatment of the hydrochar to remove phytotoxic compounds and increase its carbon content. This is used when targeting the land application market for hydrochar. The annual OPEX was in the region of £1.2 million for each of the feedstocks. The CAPEX and OPEX were validated by industrial data and were found to be in good agreement. This offers an improvement over previous literature estimates, which were shown to give significant under-estimates for both energy use and costs. The effect of plant capacity on the costs was then investigated. The maximum volume of feedstock each HTC reactor could hold was set to 8 m^3, as per industrial designs. Therefore, as the plant capacity increased, so did the number of reactors. There were local minimums in terms of a trade-off between feedstock processed and CAPEX. The default plant size, treating 41,600 tonnes/year of wet feedstock, was the maximum that a 2-reactor system could process. As the plant size and number of reactors increased, savings were available due to economies of scale. However, an important consideration for the waste treatments is the availability and location of the waste itself. This idea was specifically investigated as part of a case study of the HTC of digestate in the UK.

The process model was run under a range of operating conditions to investigate optimisation. Unsurprisingly, lower HTC temperatures and residence times led to a decrease in OPEX due to a reduction in thermal energy demand, and CAPEX as shorter residence times reduced the required reactor size. Also, as per the experimental findings, less severe HTC conditions increased the yield of hydrochar. However, it also led to a decrease in the carbon content and thus energy content of the hydrochar. When considering the optimisation of the plant, an operator will be interested in not only the plant costs, but also the potential profits from product sales. Therefore, a series of investment analyses using different assumptions about product markets were performed for a range of feedstocks.

7.4 Potential for HTC in the UK market

When looking at the potential for HTC technology in the UK, a number of important factors were considered: availability and current management of feedstock, HTC products and their markets, plant logistics, and future changes in policy or opinion. The majority of this analysis was based on a case-study of the HTC of digestate in the UK. Digestate is a suitable candidate for HTC because of its high moisture content.

Digestate generation in the UK has been rising since the onset of AD plants and while there are existing digestate management practices, these are costly and at risk of more stringent restrictions on its use. Despite this, digestate has been highlighted as a potentially valuable material. Each year, 10.9 million tonnes of whole digestate is produced, equivalent to 500 thousand tonnes on a dry basis, and contains 133–240 thousand tonnes of carbon as well as high quantities of plant available nutrients. Large, commercial and industrial-scale AD operators often find themselves having to pay for the removal of digestate. Whole digestate has a very high moisture content, often up to 97 wt.%, which results in large transportation costs. To reduce this, dewatering technology can be implemented, resulting in a digestate fibre with a moisture content of roughly 75 wt.% and digestate liquor. This is common practice in wastewater treatment plants treating sewage sludge digestate, but it is still a developing technology for other forms of digestate that are more complicated to dewater. The cost of transporting and spreading to land was estimated at £16/tonne of digestate fibre. Therefore, it was conceived that this cost could be saved if digestate were diverted to HTC instead.

The location and availability of waste is important when considering waste treatments and so transportation costs to a central HTC plant were estimated. Both Nottinghamshire and Lincolnshire, used as examples, showed similar relationships in the trade-off between HTC plant capacity and transportation costs. A 2-reactor HTC plant treating 41,600 tonnes of wet feedstock a year resulted in the lowest cost option per tonne of feedstock processed. At a larger scale, any savings due to economies of scale were offset by the required increase in transportation distance and cost.

To evaluate the overall economic potential of a HTC plant, the CAPEX, OPEX, savings compared to current digestate management, and HTC product sales were taken into account. Hydrochar as a solid fuel has a predicted sale price of somewhere in the region of £150/tonne. From the experimental results, it was found that hydrochar from the crop-based digestate was most suitable as a solid fuel. The hydrochars from waste-based and farm-based digestates, however, had problematic high ash contents. But their higher nutrient contents meant they showed more promise as a fertiliser or for carbon sequestration. Based on the elemental composition, the predicted hydrochar sale price from the waste-based digestate came to

£64/tonne for a fertiliser and £89/tonne for carbon sequestration. As the farm-based digestate had a higher nutrient and carbon content, its sale prices came to £79/tonne for a fertiliser and £134/tonne for carbon sequestration. However, it is important to note, that if hydrochar is produced for use in carbon sequestration, the CAPEX and OPEX of the plant will increase due to the additional thermal post-treatment stage required.

An investment analysis was performed for all three types of digestate over a 20-year period. As the crop-based digestate resulted in a higher yield and potential market price of hydrochar, it had the shortest payback period of 13 years. The payback period of the other types of digestate exceeded this, with roughly 15 years for the farm-based digestate and in excess of 20 years for the waste-based digestate. In industry, shorter payback periods are desired and so the sale of the process liquid as a fertiliser was also considered. Based upon the nutrient contents of the experimental samples, the sale price of the waste-based process liquid was estimated at £5/tonne, and the farm-based sample at £6/tonne. The sale of such process liquid reduced the payback period of the HTC plant by over 5 years. In these calculations, it was assumed that no further processing of the liquid wase required prior to being spread as a fertiliser. There is no common practice across HTC plant operators for the management of process liquid currently. Therefore, the model was kept simple, but it is likely that the spreading of process liquid at industrial scale would incur treatment costs which have not been accounted for.

For comparison, investment analyses were also performed for two lignocellulosic biomasses: miscanthus and wood pellets. The process model was run for these feedstocks with yield and hydrochar properties defined by experimental work previously carried out using the same HTC test rig. While the lignocellulosic biomasses resulted in a higher yield and higher quality hydrochar, the increase in hydrochar sales did not offset the increased feedstock cost. Rather than offering savings of £16/tonne of feedstock used, the lignocellulosic feedstocks came at a cost of £50–80/tonne. Therefore, over a 20-year period, the investment analysis showed a negative NPV and therefore a steady decrease in profits year upon year, proving to be financial unviable.

As the digestate and hydrochar product markets are developing, a sensitivity analysis was performed to find what sale prices and/or savings over current digestate management costs were required to achieve a defined payback period. With a digestate saving of £16/tonne, hydrochar sale prices of £200–300/tonne were required for a 10-year payback period. Alternatively, with the hydrochar sale price fixed at £150/tonne, savings of £30–40/tonne of digestate fibre were required to meet a 10-year payback period. Note that neither of these scenarios included the extra revenue from the sale of process liquid as its market is less

established. While the HTC of digestate in the UK does not appear to be a financially viable option in the current climate, this analysis clearly indicates under what conditions it would become so. The accuracy and applicability of these validated results are a large improvement on what is available in existing literature. They also offer an in-depth understanding of the technical and financial feasibility of using HTC to treat digestate, which is important as the quantity of digestate produced in the UK increases. As the management costs of digestate are expected to rise, this is valuable information for the future as operators look to alternative management options.

7.5 Future work

The purpose of this work was to understand the technical and economic feasibility of HTC, using UK digestate as an example feedstock. A limited number of experimental runs were performed to create regression models. If there was interest in a particular market for HTC products, further experimental work is recommended. For example, there was some disparity in the measured elemental concentrations, particularly nitrogen, in the hydrochar and process liquid. Therefore, in order to gain a better understanding of the potential of either product for the fertiliser market, the nutrient contents of both should be measured using sensitive analysis equipment. If the focus is on using hydrochar for carbon sequestration, the permanence of carbon in the soil should be studied. There are currently only a few long-term field experiments considering hydrochar, which have conflicting results. The feedstock, HTC conditions and use of various post-treatments all have an effect on the stability of hydrochar. A robust, comprehensive field experiment of multiple types of hydrochar would be of real benefit.

This work has focussed on the HTC of digestate, but not all integration opportunities with the AD plant have been studied. For example, supplying a portion of the thermal heat required by HTC with the waste heat from the AD plant could cut operating costs as well as GHG emissions. Additionally, the suitability of using the HTC process liquid as a substrate for AD could also be considered in greater depth. In theory, a truly integrated AD and HTC plant would cut transportation costs and could prove economically feasible when located at a large-scale AD plant.

While digestate as a HTC feedstock is of interest in the UK, a case-study of the financial potential of other feedstocks in alternative locations is recommended for a clearer understanding of the local market. Further experimental runs using that feedstock could also be useful, especially if it were significantly different in properties to digestate. It is suggested that the techno-economics are taken into account when choosing conditions for experimental runs. This would mean the results were more targeted and similar to the expected industrial conditions.

Overall, this work has set out a good framework for exploring the potential of HTC and what thresholds need to be met before it becomes economically feasible. For a more in-depth study of a particular feedstock or business model, further work is recommended. This could involve experimental runs, modelling the integration of processes or a more targeted review of the relevant current markets.

8 References

1. Pan, S.Y.; Du, M.A.; Huang, I. Te; Liu, I.H.; Chang, E.E.; Chiang, P.C. Strategies on implementation of waste-to-energy (WTE) supply chain for circular economy system: a review. *J. Clean. Prod.* **2015**, *108*, 409–421, doi:10.1016/j.jclepro.2015.06.124.

2. Attorp, A.; McAreavey, R. Muck, brass and smoke: Policy post-exceptionalism in the agri-food sector. *J. Rural Stud.* **2020**, *79*, 302–310, doi:10.1016/j.jrurstud.2020.08.050.

3. Garlapalli, R.K.; Wirth, B.; Reza, M.T. Pyrolysis of hydrochar from digestate: Effect of hydrothermal carbonization and pyrolysis temperatures on pyrochar formation. *Bioresour. Technol.* **2016**, *220*, 168–174, doi:10.1016/j.biortech.2016.08.071.

4. Lu, X.; Jordan, B.; Berge, N.D. Thermal conversion of municipal solid waste via hydrothermal carbonization: Comparison of carbonization products to products from current waste management techniques. *Waste Manag.* **2012**, *32*, 1353–1365, doi:10.1016/j.wasman.2012.02.012.

5. Reza, M.T.; Coronella, C.; Holtman, K.M.; Franqui-Villanueva, D.; Poulson, S.R. Hydrothermal Carbonization of Autoclaved Municipal Solid Waste Pulp and Anaerobically Treated Pulp Digestate. *ACS Sustain. Chem. Eng.* **2016**, *4*, 3649–3658, doi:10.1021/acssuschemeng.6b00160.

6. DEFRA *UK Statistics on Waste*; 2020;

7. Tuck, C.O.; Perez, E.; Horvath, I.T.; Sheldon, R.A.; Poliakoff, M. Valorization of Biomass: Deriving More Value from Waste. *Science (80-.).* **2012**, *337*, 695–699, doi:10.1126/science.1218930.

8. House of Lords Science and Technology Select Committee *Waste or resource? Stimulating a bioeconomy*; Authority of the House of Lords: London : The Stationery Office Limited, 2014;

9. Zero Waste Scotland *The composition of municipal solid waste in Scotland*; 2010;

10. Defra *Municipal Waste Composition: A Review of Municipal Waste Component Analyses*; 2009;

11. The European Parliament and The Council of the European Union *on waste and repealing certain Directives. 2008/98/EC*; 2008;

12. European Council *on the landfill of waste. 1999/31/EC*; 1999; p. L182/1-19;

13. Department for Business Innovation & Skills *Building a high value bioeconomy: opportunities from waste*; Department for Business, I.& S., Ed.; 2015;

14. Cao, Y.; Pawłowski, A. Sewage sludge-to-energy approaches based on anaerobic digestion and pyrolysis: Brief overview and energy efficiency assessment. *Renew. Sustain. Energy Rev.* **2012**, *16*, 1657–1665, doi:10.1016/j.rser.2011.12.014.

15. NNFCC The Anaerobic Digestion (AD) Portal - Biogas Map Available online: http://www.biogas-info.co.uk/resources/biogas-map/ (accessed on Oct 16, 2019).

16. WRAP *Organics recycling industry status report 2015*; 2015;

17. WRAP *DC-Agri; field experiments for quality digestate and compost in agriculture*; 2016;

18. Liu, J.; Huang, S.; Chen, K.; Wang, T.; Mei, M.; Li, J. Preparation of biochar from food waste digestate: Pyrolysis behavior and product properties. *Bioresour. Technol.* **2020**, *302*, 122841, doi:10.1016/j.biortech.2020.122841.

19. NNFCC *Anaerobic Digestion Deployment in the UK Seventh Annual Report*; 2020;

20. WRAP Cymru *Assessing the Costs and Benefits for Production and Beneficial Application of Anaerobic Digestate to Agricultural Land in Wales*; 2014;

21. WRAP *Digestate and compost use in agriculture: Good practice guidance*; 2016;

22. BMNT *Studie Über die Erschließung des Potenzials biogener Haushaltsabfälle und Grünschnitt zum Zwecke der Verwertung in einer Biogasanlage zur optimierten energetischen und stofflichen Verwertung*; 2011;

23. WRAP *Digestate distribution models*; 2013;

24. NNFCC *Assessment of digestate drying as an eligible heat use in the Renewable Heat Incentive*; 2016;

25. Libra, J.A.; Ro, K.S.; Kammann, C.; Funke, A.; Berge, N.D.; Neubauer, Y.; Titirici, M.-M.; Fühner, C.; Bens, O.; Kern, J.; et al. Hydrothermal carbonization of biomass residuals: a comparative review of the chemistry, processes and applications of wet and dry pyrolysis. *Biofuels* **2011**, *2*, 71–106, doi:10.4155/bfs.10.81.

26. Bergius, F. *Die Anwendung hoher Drucke bei chemischen Vorgängen und eine Nachbildung des Entstehungsprozesses der Steinkohle*; Halle a. S. Knapp, 1913;

27. Hilber, B. Method and device for hydrothermal carbonisation of biomass. EP1970431B1. European Patent Office 2008.

28. Antonietti, M.; Eisner, P.; Malberg, A.; Menner, M.; Staebler, A. Method for the wet-chemical transformation of biomass by hydrothermal carbonization. ZA200906001B. Companies and Intellectual Property Commission 2008.

29. Hofer, L. Hydrothermal carbonization of biomass. EP2131953A1. European Patent Office 2008.

30. Funke, Axel; Ziegler, F. Hydrothermal carbonization of biomass: A summary and discussion of chemical mechanisms for process engineering. *Biofuels, Bioprod. Biorefining* **2010**, *4*, 160–177, doi:10.1002/bbb.198.

31. Hoekman, S.K.; Broch, A.; Felix, L.; Farthing, W. Hydrothermal carbonization (HTC) of loblolly pine using a continuous, reactive twin-screw extruder. *Energy Convers. Manag.* **2017**, *134*, 247–259, doi:10.1016/j.enconman.2016.12.035.

32. Sevilla, M.; Fuertes, A.B. The production of carbon materials by hydrothermal carbonization of cellulose. *Carbon N. Y.* **2009**, *47*, 2281–2289, doi:10.1016/j.carbon.2009.04.026.

33. Lei, Y.; Su, H.; Tian, R. Morphology evolution, formation mechanism and adsorption properties of hydrochars prepared by hydrothermal carbonization of corn stalk. *RSC Adv.* **2016**, *6*, 107829–107835, doi:10.1039/c6ra21607b.

34. Berge, N.D.; Ro, K.S.; Mao, J.; Flora, J.R. V; Chappell, M.A.; Bae, S. Hydrothermal Carbonization of Municipal Waste Streams. *Environ. Sci. Technol.* **2011**, *45*, 5696–5703, doi:10.1021/es2004528.

35. Pecchi, M.; Patuzzi, F.; Basso, D.; Baratieri, M. Enthalpy change during hydrothermal carbonization of biomass: a critical review. *J. Therm. Anal. Calorim.* **2019**, doi:10.1007/s10973-019-09117-4.

36. Spinosa, L.; Ayol, A.; Baudez, J.-C.; Canziani, R.; Jenicek, P.; Leonard, A.; Rulkens, W.; Xu, G.; Van Dijk, L. Sustainable and Innovative Solutions for Sewage Sludge Management. *Water* **2011**, *3*, 702–717.

37. Conti, R.; Rombolà, A.G.; Modelli, A.; Torri, C.; Fabbri, D. Evaluation of the thermal and environmental stability of switchgrass biochars by Py-GC-MS. *J. Anal. Appl. Pyrolysis* **2014**, *110*, 239–247, doi:10.1016/j.jaap.2014.09.010.

38. Fonts, I.; Azuara, M.; Gea, G.; Murillo, M.B. Study of the pyrolysis liquids obtained from different sewage sludge. *J. Anal. Appl. Pyrolysis* **2009**, *85*, 184–191, doi:10.1016/j.jaap.2008.11.003.

39. Affam, A.C.; Ezechi, E.H. *Handbook of Research on Resource Management for Pollution and Waste Treatment*; IGI Global, 2020; ISBN 978-1-79980-369-0.

40. Suwelack, K.U.; Wüst, D.; Fleischmann, P.; Kruse, A. Prediction of gaseous, liquid and solid mass yields from hydrothermal carbonization of biogas digestate by severity parameter. *Biomass Convers. Biorefinery* **2016**, *6*, 151–160, doi:10.1007/s13399-015-0172-8.

41. Lynam, J.G.; Reza, M.T.; Yan, W.; Vásquez, V.R.; Coronella, C.J. Hydrothermal carbonization of various lignocellulosic biomass. *Biomass Convers. Biorefinery* **2015**, *5*, 173–181, doi:10.1007/s13399-014-0137-3.

42. Danso-Boateng, E.; Shama, G.; Wheatley, A.D.; Martin, S.J.; Holdich, R.G. Hydrothermal carbonisation of sewage sludge: Effect of process conditions on product characteristics and methane production. *Bioresour. Technol.* **2015**, *177*, 318–327, doi:10.1016/j.biortech.2014.11.096.

43. Mumme, J.; Eckervogt, L.; Pielert, J.; Diakité, M.; Rupp, F.; Kern, J. Hydrothermal carbonization of anaerobically digested maize silage. *Bioresour. Technol.* **2011**, *102*, 9255–9260, doi:10.1016/j.biortech.2011.06.099.

44. Toptas Tag, A.; Duman, G.; Yanik, J. Influences of feedstock type and process variables on hydrochar properties. *Bioresour. Technol.* **2018**, *250*, 337–344, doi:10.1016/j.biortech.2017.11.058.

45. Smith, A.M.; Ross, A.B. The Influence of Residence Time during Hydrothermal Carbonisation of Miscanthus on Bio-Coal Combustion Chemistry. *Energies* **2019**, *12*, 523, doi:10.3390/en12030523.

46. Tasca, A.L.; Puccini, M.; Gori, R.; Corsi, I.; Galletti, A.M.R.; Vitolo, S. Hydrothermal carbonization of sewage sludge: A critical analysis of process severity, hydrochar properties and environmental implications. *Waste Manag.* **2019**, *93*, 1–13, doi:10.1016/j.wasman.2019.05.027.

47. Wilk, M.; Magdziarz, A.; Kalemba-Rec, I.; Szymańska-Chargot, M. Upgrading of green waste into carbon-rich solid biofuel by hydrothermal carbonization: The effect of process parameters on hydrochar derived from acacia. *Energy* **2020**, *202*,

doi:10.1016/j.energy.2020.117717.

48. Yan, W.; Hoekman, S.K.; Broch, A.; Coronella, C.J. Effect of hydrothermal carbonization reaction parameters on the properties of hydrochar and pellets. *Environ. Prog. Sustain. Energy* **2014**, *33*, 676–680, doi:10.1002/ep.11974.

49. Benavente, V.; Calabuig, E.; Fullana, A. Upgrading of moist agro-industrial wastes by hydrothermal carbonization. *J. Anal. Appl. Pyrolysis* **2015**, *113*, 89–98, doi:10.1016/j.jaap.2014.11.004.

50. Mäkelä, M.; Benavente, V.; Fullana, A. Hydrothermal carbonization of lignocellulosic biomass: Effect of process conditions on hydrochar properties. *Appl. Energy* **2015**, *155*, 576–584, doi:10.1016/j.apenergy.2015.06.022.

51. Basso, D.; Patuzzi, F.; Castello, D.; Baratieri, M.; Rada, E.C.; Weiss-Hortala, E.; Fiori, L. Agro-industrial waste to solid biofuel through hydrothermal carbonization. *Waste Manag.* **2016**, *47*, 114–121, doi:10.1016/j.wasman.2015.05.013.

52. Sabio, E.; Álvarez-Murillo, A.; Román, S.; Ledesma, B. Conversion of tomato-peel waste into solid fuel by hydrothermal carbonization: Influence of the processing variables. *Waste Manag.* **2016**, *47*, 122–132, doi:10.1016/j.wasman.2015.04.016.

53. Nizamuddin, S.; Baloch, H.A.; Griffin, G.J.; Mubarak, N.M.; Bhutto, A.W.; Abro, R.; Mazari, S.A.; Ali, B.S. An overview of effect of process parameters on hydrothermal carbonization of biomass. *Renew. Sustain. Energy Rev.* **2017**, *73*, 1289–1299, doi:10.1016/j.rser.2016.12.122.

54. Guo, S.; Dong, X.; Wu, T.; Zhu, C. Influence of reaction conditions and feedstock on hydrochar properties. *Energy Convers. Manag.* **2016**, *123*, 95–103, doi:10.1016/j.enconman.2016.06.029.

55. Volpe, M.; Fiori, L. From olive waste to solid biofuel through hydrothermal carbonisation: The role of temperature and solid load on secondary char formation and hydrochar energy properties. *J. Anal. Appl. Pyrolysis* **2017**, *124*, 63–72, doi:10.1016/j.jaap.2017.02.022.

56. Stutzenstein, P.; Bacher, M.; Rosenau, T.; Pfeifer, C. Optimization of Nutrient and Carbon Recovery from Anaerobic Digestate via Hydrothermal Carbonization and Investigation of the Influence of the Process Parameters. *Waste and Biomass Valorization* **2018**, *9*, 1303–1318, doi:10.1007/s12649-017-9902-4.

57. Smith, A.M.; Ekpo, U.; Ross, A.B. The influence of pH on the combustion properties of bio-coal following hydrothermal treatment of swine manure. *Energies* **2020**, *13*, 1–20, doi:10.3390/en13020331.

58. Li, L.; Wang, Y.; Xu, J.; Flora, J.R.V.; Hoque, S.; Berge, N.D. Quantifying the sensitivity of feedstock properties and process conditions on hydrochar yield, carbon content, and energy content. *Bioresour. Technol.* **2018**, *262*, 284–293, doi:10.1016/j.biortech.2018.04.066.

59. Overend, R.P.; Chornet, E. Fractionation of lignocellulosics by steam-aqueous pretreatments. *Philos. Trans. R. Soc. London. Ser. A, Math. Phys. Sci.* **1987**, *321*, 523–536, doi:10.1098/rsta.1987.0029.

60. Jeder, A.; Sanchez-Sanchez, A.; Gadonneix, P.; Masson, E.; Ouederni, A.; Celzard, A.; Fierro, V. The severity factor as a useful tool for producing hydrochars and derived carbon materials. *Environ. Sci. Pollut. Res.* **2017**, doi:10.1007/s11356-017-0366-7.

61. Abatzoglou, N.; Chornet, E.; Belkacemi, K.; Overend, R.P. Phenomenological kinetics of complex systems: the development of a generalized severity parameter and its application to lignocellulosics fractionation. *Chem. Eng. Sci.* **1992**, *47*, 1109–1122, doi:10.1016/0009-2509(92)80235-5.

62. Volpe, M.; Goldfarb, J.L.; Fiori, L. Hydrothermal carbonization of Opuntia ficus-indica cladodes: Role of process parameters on hydrochar properties. *Bioresour. Technol.* **2017**, *247*, 310–318, doi:10.1016/j.biortech.2017.09.072.

63. Álvarez-Murillo, A.; Sabio, E.; Ledesma, B.; Román, S.; González-García, C.M. Generation of biofuel from hydrothermal carbonization of cellulose. Kinetics modelling. *Energy* **2016**, *94*, 600–608, doi:10.1016/j.energy.2015.11.024.

64. Heidari, M.; Norouzi, O.; Salaudeen, S.; Acharya, B.; Dutta, A. Prediction of Hydrothermal Carbonization with Respect to the Biomass Components and Severity Factor. *Energy & Fuels* **2019**, *33*, 9916–9924, doi:10.1021/acs.energyfuels.9b02291.

65. Li, L.; Flora, J.R. V; Caicedo, J.M.; Berge, N.D. Investigating the role of feedstock properties and process conditions on products formed during the hydrothermal carbonization of organics using regression techniques. *Bioresour. Technol.* **2015**, *187*, 263–274, doi:10.1016/j.biortech.2015.03.054.

66. Rutherford, D.W.; Chiou, C.T.; Kile, D.E. Influence of Soil Organic Matter Composition on the Partition of Organic Compounds. *Environ. Sci. Technol.* **1992**, *26*, 336–340,

doi:10.1021/es00026a014.

67. Vallejo, F.; Díaz-Robles, L.A.; Vega, R.; Cubillos, F. A novel approach for prediction of mass yield and higher calorific value of hydrothermal carbonization by a robust multilinear model and regression trees. *J. Energy Inst.* **2020**, *93*, 1755–1762, doi:10.1016/j.joei.2020.03.006.

68. Belkacemi, K.; Abatzoglou, N.; Overend, R.P.; Chornet, E. Phenomenological kinetics of complex systems: mechanistic considerations in the solubilization of hemicelluloses following aqueous/steam treatments. *Ind. Eng. Chem. Res.* **1991**, *30*, 2416–2425, doi:10.1021/ie00059a009.

69. Leger, S.; Chornet, E.; Overend, R.P. Characterization and quantification of changes occurring in the low-severity dewatering of peat. *Int. J. Coal Geol.* **1987**, *8*, 135–146, doi:10.1016/0166-5162(87)90028-0.

70. Stirling, R.J.; Snape, C.E.; Meredith, W. The impact of hydrothermal carbonisation on the char reactivity of biomass. *Fuel Process. Technol.* **2018**, *177*, 152–158, doi:10.1016/j.fuproc.2018.04.023.

71. Akarsu, K.; Duman, G.; Yilmazer, A.; Keskin, T.; Azbar, N.; Yanik, J. Sustainable valorization of food wastes into solid fuel by hydrothermal carbonization. *Bioresour. Technol.* **2019**, *292*, 121959, doi:10.1016/j.biortech.2019.121959.

72. Koppejan, J.; van Loo, S. *The Handbook of Biomass Combustion and Co-firing*; Routledge, 2012;

73. Smith, A.M.; Singh, S.; Ross, A.B. Fate of inorganic material during hydrothermal carbonisation of biomass: Influence of feedstock on combustion behaviour of hydrochar. *Fuel* **2016**, *169*, 135–145, doi:10.1016/j.fuel.2015.12.006.

74. Smith, A.M.; Whittaker, C.; Shield, I.; Ross, A.B. The potential for production of high quality bio-coal from early harvested Miscanthus by hydrothermal carbonisation. *Fuel* **2018**, *220*, 546–557, doi:10.1016/j.fuel.2018.01.143.

75. He, C.; Giannis, A.; Wang, J.Y. Conversion of sewage sludge to clean solid fuel using hydrothermal carbonization: Hydrochar fuel characteristics and combustion behavior. *Appl. Energy* **2013**, *111*, 257–266, doi:10.1016/j.apenergy.2013.04.084.

76. Zhuang, X.; Zhan, H.; Huang, Y.; Song, Y.; Yin, X.; Wu, C. Conversion of industrial biowastes to clean solid fuels via hydrothermal carbonization (HTC): Upgrading

mechanism in relation to coalification process and combustion behavior. *Bioresour. Technol.* **2018**, *267*, 17–29, doi:10.1016/j.biortech.2018.07.002.

77. Wang, R.; Wang, C.; Zhao, Z.; Jia, J.; Jin, Q. Energy recovery from high-ash municipal sewage sludge by hydrothermal carbonization: Fuel characteristics of biosolid products. *Energy* **2019**, *186*, 115848, doi:10.1016/j.energy.2019.07.178.

78. Mazumder, S.; Saha, P.; Reza, M.T. Co-hydrothermal carbonization of coal waste and food waste: fuel characteristics. *Biomass Convers. Biorefinery* **2020**, doi:10.1007/s13399-020-00771-5.

79. Sevilla, M.; Maciá-Agulló, J.A.; Fuertes, A.B. Hydrothermal carbonization of biomass as a route for the sequestration of CO2: Chemical and structural properties of the carbonized products. *Biomass and Bioenergy* **2011**, *35*, 3152–3159, doi:10.1016/j.biombioe.2011.04.032.

80. Bargmann, I.; Rillig, M.C.; Buss, W.; Kruse, A.; Kuecke, M. Hydrochar and biochar effects on germination of spring barley. *J. Agron. Crop Sci.* **2013**, *199*, 360–373, doi:10.1111/jac.12024.

81. Busch, D.; Stark, A.; Kammann, C.I.; Glaser, B. Genotoxic and phytotoxic risk assessment of fresh and treated hydrochar from hydrothermal carbonization compared to biochar from pyrolysis. *Ecotoxicol. Environ. Saf.* **2013**, *97*, 59–66, doi:10.1016/j.ecoenv.2013.07.003.

82. Zhu, X.; Liu, Y.; Qian, F.; Zhang, S.; Chen, J. Investigation on the Physical and Chemical Properties of Hydrochar and Its Derived Pyrolysis Char for Their Potential Application: Influence of Hydrothermal Carbonization Conditions. *Energy & Fuels* **2015**, *29*, 5222–5230, doi:10.1021/acs.energyfuels.5b00512.

83. Fornes, F.; Belda, R.M. Biochar versus hydrochar as growth media constituents for ornamental plant cultivation. *Sci. Agric.* **2018**, *75*, 304–312, doi:10.1590/1678-992x-2017-0062.

84. Roehrdanz, M.; Greve, T.; de Jager, M.; Buchwald, R.; Wark, M. Co-composted hydrochar substrates as growing media for horticultural crops. *Sci. Hortic. (Amsterdam).* **2019**, *252*, 96–103, doi:10.1016/j.scienta.2019.03.055.

85. Álvarez, M.L.; Gascó, G.; Plaza, C.; Paz-Ferreiro, J.; Méndez, A. Hydrochars from Biosolids and Urban Wastes as Substitute Materials for Peat. *L. Degrad. Dev.* **2017**, doi:10.1002/ldr.2756.

86. Hitzl, M.; Mendez, A.; Owsianiak, M.; Renz, M. Making hydrochar suitable for agricultural soil: A thermal treatment to remove organic phytotoxic compounds. *J. Environ. Chem. Eng.* **2018**, *6*, 7029–7034, doi:10.1016/j.jece.2018.10.064.

87. Bahcivanji, L.; Gascó, G.; Paz-Ferreiro, J.; Méndez, A. The effect of post-pyrolysis treatment on waste biomass derived hydrochar. *Waste Manag.* **2020**, *106*, 55–61, doi:10.1016/j.wasman.2020.03.007.

88. Mumme, J.; Getz, J.; Prasad, M.; Lüder, U.; Kern, J.; Mašek, O.; Buss, W. Toxicity screening of biochar-mineral composites using germination tests. *Chemosphere* **2018**, *207*, 91–100, doi:10.1016/j.chemosphere.2018.05.042.

89. Dalias, P.; Prasad, M.; Mumme, J.; Kern, J.; Stylianou, M.; Christou, A. Low-cost post-treatments improve the efficacy of hydrochar as peat replacement in growing media. *J. Environ. Chem. Eng.* **2018**, *6*, 6647–6652, doi:10.1016/j.jece.2018.10.042.

90. Melo, T.M.; Bottlinger, M.; Schulz, E.; Leandro, W.M.; Botelho de Oliveira, S.; Menezes de Aguiar Filho, A.; El-Naggar, A.; Bolan, N.; Wang, H.; Ok, Y.S.; et al. Management of biosolids-derived hydrochar (Sewchar): Effect on plant germination, and farmers' acceptance. *J. Environ. Manage.* **2019**, *237*, 200–214, doi:10.1016/j.jenvman.2019.02.042.

91. International Biochar Initiative *Standardized Product Definition and Product Testing Guidelines for Biochar That Is Used in Soil*; 2015;

92. EBC *European Biochar Certificate - Guidelines for a Sustainable Production of Biochar*; 2019;

93. Department for Environment Food & Rural Affairs *Sewage sludge in agriculture: code of practice for England, Wales and Northern Ireland*; 2018;

94. Miliotti, E.; Casini, D.; Rosi, L.; Lotti, G.; Rizzo, A.M.; Chiaramonti, D. Lab-scale pyrolysis and hydrothermal carbonization of biomass digestate: Characterization of solid products and compliance with biochar standards. *Biomass and Bioenergy* **2020**, *139*, 105593, doi:10.1016/j.biombioe.2020.105593.

95. Lanza, G.; Stang, A.; Kern, J.; Wirth, S.; Gessler, A. Degradability of raw and post-processed chars in a two-year field experiment. *Sci. Total Environ.* **2018**, *628–629*, 1600–1608, doi:10.1016/j.scitotenv.2018.02.164.

96. Gronwald, M.; Vos, C.; Helfrich, M.; Don, A. Stability of pyrochar and hydrochar in

agricultural soil - a new field incubation method. *Geoderma* **2016**, *284*, 85–92, doi:10.1016/j.geoderma.2016.08.019.

97. Idowu, I.; Li, L.; Flora, J.R.V.; Pellechia, P.J.; Darko, S.A.; Ro, K.S.; Berge, N.D. Hydrothermal carbonization of food waste for nutrient recovery and reuse. *Waste Manag.* **2017**, *69*, 480–491, doi:10.1016/j.wasman.2017.08.051.

98. Hitzl, M.; Corma, A.; Pomares, F.; Renz, M. The hydrothermal carbonization (HTC) plant as a decentral biorefinery for wet biomass. *Catal. Today* **2014**, *257*, 154–159, doi:10.1016/j.cattod.2014.09.024.

99. Uddin, M.H.; Reza, M.T.; Lynam, J.G.; Coronella, C.J. Effects of water recycling in hydrothermal carbonization of loblolly pine. *Environ. Prog. Sustain. Energy* **2013**, *33*, 1309–1315, doi:10.1002/ep.11899.

100. Kabadayi Catalkopru, A.; Kantarli, I.C.; Yanik, J. Effects of spent liquor recirculation in hydrothermal carbonization. *Bioresour. Technol.* **2017**, *226*, 89–93, doi:10.1016/j.biortech.2016.12.015.

101. Chen, X.; Ma, X.; Peng, X.; Lin, Y.; Wang, J.; Zheng, C. Effects of aqueous phase recirculation in hydrothermal carbonization of sweet potato waste. *Bioresour. Technol.* **2018**, *267*, 167–174, doi:10.1016/j.biortech.2018.07.032.

102. Kambo, H.S.; Minaret, J.; Dutta, A. Process Water from the Hydrothermal Carbonization of Biomass: A Waste or a Valuable Product? *Waste and Biomass Valorization* **2018**, *9*, 1181–1189, doi:10.1007/s12649-017-9914-0.

103. Köchermann, J.; Görsch, K.; Wirth, B.; Mühlenberg, J.; Klemm, M. Hydrothermal carbonization: Temperature influence on hydrochar and aqueous phase composition during process water recirculation. *J. Environ. Chem. Eng.* **2018**, *6*, 5481–5487, doi:10.1016/J.JECE.2018.07.053.

104. Wang, F.; Wang, J.; Gu, C.; Han, Y.; Zan, S.; Wu, S. Effects of process water recirculation on solid and liquid products from hydrothermal carbonization of Laminaria. *Bioresour. Technol.* **2019**, *292*, 121996, doi:10.1016/j.biortech.2019.121996.

105. Wirth, B.; Mumme, J. Anaerobic Digestion of Waste Water from Hydrothermal Carbonization of Corn Silage. *Appl. Bioenergy* **2014**, *1*, 1–10, doi:10.2478/apbi-2013-0001.

106. Pagés-Díaz, J.; Cerda Alvarado, A.O.; Montalvo, S.; Diaz-Robles, L.; Curio, C.H.

Anaerobic bio-methane potential of the liquors from hydrothermal carbonization of different lignocellulose biomasses. *Renew. Energy* **2020**, *157*, 182–189, doi:10.1016/j.renene.2020.05.025.

107. Villamil, J.A.; Mohedano, A.F.; Rodriguez, J.J.; de la Rubia, M.A. Valorisation of the liquid fraction from hydrothermal carbonisation of sewage sludge by anaerobic digestion. *J. Chem. Technol. Biotechnol.* **2018**, *93*, 450–456, doi:10.1002/jctb.5375.

108. Chen, H.; Rao, Y.; Cao, L.; Shi, Y.; Hao, S.; Luo, G.; Zhang, S. Hydrothermal conversion of sewage sludge: Focusing on the characterization of liquid products and their methane yields. *Chem. Eng. J.* **2019**, *357*, 367–375, doi:10.1016/j.cej.2018.09.180.

109. Marin-Batista, J.D.; Villamil, J.A.; Rodriguez, J.J.; Mohedano, A.F.; de la Rubia, M.A. Valorization of microalgal biomass by hydrothermal carbonization and anaerobic digestion. *Bioresour. Technol.* **2018**, *274*, 395–402, doi:10.1016/j.biortech.2018.11.103.

110. Parmar, K.R.; Ross, A.B. Integration of Hydrothermal Carbonisation with Anaerobic Digestion; Opportunities for Valorisation of Digestate. *Energies* **2019**, *12*, 1586, doi:10.3390/en12091586.

111. Zhao, K.; Li, Y.; Zhou, Y.; Guo, W.; Jiang, H.; Xu, Q. Characterization of hydrothermal carbonization products (hydrochars and spent liquor) and their biomethane production performance. *Bioresour. Technol.* **2018**, *267*, 9–16, doi:10.1016/j.biortech.2018.07.006.

112. Brown, A.E.; Finnerty, G.L.; Camargo-Valero, M.A.; Ros, A.B. Valorisation of macroalgae via the integration of hydrothermal carbonisation and anaerobic digestion. *Bioresour. Technol.* **2020**, *312*, 123539, doi:10.1016/j.biortech.2020.123539.

113. Becker, R.; Dorgerloh, U.; Paulke, E.; Mumme, J.; Nehls, I. Hydrothermal Carbonization of Biomass: Major Organic Components of the Aqueous Phase. *Chem. Eng. Technol.* **2014**, *37*, 511–518, doi:10.1002/ceat.201300401.

114. Fregolente, L.G.; Miguel, T.B.A.R.; de Castro Miguel, E.; de Almeida Melo, C.; Moreira, A.B.; Ferreira, O.P.; Bisinoti, M.C. Toxicity evaluation of process water from hydrothermal carbonization of sugarcane industry by-products. *Environ. Sci. Pollut. Res.* **2018**, doi:10.1007/s11356-018-1771-2.

115. Reza, M.T.; Freitas, A.; Yang, X.; Coronella, C.J. Wet Air Oxidation of Hydrothermal Carbonization (HTC) Process Liquid. *ACS Sustain. Chem. Eng.* **2016**, *4*, 3250–3254,

doi:10.1021/acssuschemeng.6b00292.

116. Belete, Y.Z.; Leu, S.; Boussiba, S.; Zorin, B.; Posten, C.; Thomsen, L.; Wang, S.; Gross, A.; Bernstein, R. Characterization and utilization of hydrothermal carbonization aqueous phase as nutrient source for microalgal growth. *Bioresour. Technol.* **2019**, *290*, 121758, doi:10.1016/j.biortech.2019.121758.

117. Crossley, O.P.; Thorpe, R.B.; Lee, J. Phosphorus recovery from process waste water made by the hydrothermal carbonisation of spent coffee grounds. *Bioresour. Technol.* **2020**, *301*, 122664, doi:10.1016/j.biortech.2019.122664.

118. Mau, V.; Gross, A. Energy conversion and gas emissions from production and combustion of poultry-litter-derived hydrochar and biochar. *Appl. Energy* **2018**, *213*, 510–519, doi:10.1016/j.apenergy.2017.11.033.

119. Mäkelä, M.; Yoshikawa, K. Simulating hydrothermal treatment of sludge within a pulp and paper mill. *Appl. Energy* **2016**, *173*, 177–183, doi:10.1016/j.apenergy.2016.04.017.

120. Gupta, D.; Mahajani, S.M.; Garg, A. Effect of Hydrothermal Carbonization as Pretreatment on Energy Recovery from Food and Paper Wastes. *Bioresour. Technol.* **2019**, *285*, 121329, doi:10.1016/j.biortech.2019.121329.

121. Sharma, H.B.; Panigrahi, S.; Dubey, B.K. Hydrothermal carbonization of yard waste for solid bio-fuel production: Study on combustion kinetic, energy properties, grindability and flowability of hydrochar. *Waste Manag.* **2019**, *91*, 108–119, doi:10.1016/j.wasman.2019.04.056.

122. Danso-Boateng, E.; Holdich, R.G.; Martin, S.J.; Shama, G.; Wheatley, A.D. Process energetics for the hydrothermal carbonisation of human faecal wastes. *Energy Convers. Manag.* **2015**, *105*, 1115–1124, doi:10.1016/j.enconman.2015.08.064.

123. Zhao, P.; Shen, Y.; Ge, S.; Yoshikawa, K. Energy recycling from sewage sludge by producing solid biofuel with hydrothermal carbonization. *Energy Convers. Manag.* **2014**, *78*, 815–821, doi:10.1016/j.enconman.2013.11.026.

124. Afolabi, O.O.D.; Sohail, M.; Cheng, Y.L. Optimisation and characterisation of hydrochar production from spent coffee grounds by hydrothermal carbonisation. *Renew. Energy* **2020**, *147*, 1380–1391, doi:10.1016/j.renene.2019.09.098.

125. Department for Business Energy & Industrial Strategy *Energy Trends: UK electricity*; 2020;

126. Stemann, J.; Erlach, B.; Ziegler, F. Hydrothermal carbonisation of empty palm oil fruit bunches: Laboratory trials, plant simulation, carbon avoidance, and economic feasibility. *Waste and Biomass Valorization* **2013**, *4*, 441–454, doi:10.1007/s12649-012-9190-y.

127. Meisel, K.; Clemens, A.; Fühner, C.; Breulmann, M.; Majer, S.; Thrän, D. Comparative Life Cycle Assessment of HTC Concepts Valorizing Sewage Sludge for Energetic and Agricultural Use. *Energies* **2019**, *12*, 786, doi:10.3390/en12050786.

128. Shao, Y.; Long, Y.; Wang, H.; Liu, D.; Shen, D.; Chen, T. Hydrochar derived from green waste by microwave hydrothermal carbonization. *Renew. Energy* **2018**, doi:10.1016/j.renene.2018.09.041.

129. Nakason, K.; Panyapinyopol, B.; Kanokkantapong, V.; Viriya-empikul, N.; Kraithong, W.; Pavasant, P. Characteristics of hydrochar and hydrothermal liquid products from hydrothermal carbonization of corncob. *Biomass Convers. Biorefinery* **2017**, 199–210, doi:10.1007/s13399-017-0279-1.

130. Kempegowda, R.S.; Tran, K.-Q.; Skreiberg, Ø. Techno-economic assessment of integrated hydrochar and high-grade activated carbon production for electricity generation and storage. *Energy Procedia* **2017**, *120*, 341–348, doi:10.1016/j.egypro.2017.07.223.

131. Unrean, P.; Lai Fui, B.C.; Rianawati, E.; Acda, M. Comparative techno-economic assessment and environmental impacts of rice husk-to-fuel conversion technologies. *Energy* **2018**, *151*, 581–593, doi:10.1016/j.energy.2018.03.112.

132. Tradler, S.B.; Mayr, S.; Himmelsbach, M.; Priewasser, R.; Baumgartner, W.; Stadler, A.T. Hydrothermal carbonization as an all-inclusive process for food-waste conversion. *Bioresour. Technol. Reports* **2018**, *2*, 77–83, doi:10.1016/j.biteb.2018.04.009.

133. Saba, A.; McGaughy, K.; Reza, M. Techno-Economic Assessment of Co-Hydrothermal Carbonization of a Coal-Miscanthus Blend. *Energies* **2019**, *12*, 630, doi:10.3390/en12040630.

134. Mazumder, S.; Saha, P.; McGaughy, K.; Saba, A.; Reza, M.T. Technoeconomic analysis of co-hydrothermal carbonization of coal waste and food waste. *Biomass Convers. Biorefinery* **2020**, doi:10.1007/s13399-020-00817-8.

135. Lucian, M.; Fiori, L. Hydrothermal Carbonization of Waste Biomass: Process Design, Modeling, Energy Efficiency and Cost Analysis. *Energies* **2017**, *10*, 211,

doi:10.3390/en10020211.

136. De Mena Pardo, B.; Doyle, L.; Renz, M.; Salimbeni, A.; (Editors) *NEWAPP Industrial Scale Hydrothermal Carbonization: new applications for wet biomass waste*; 2016;

137. Román, S.; Libra, J.; Berge, N.; Sabio, E.; Ro, K.; Li, L.; Ledesma, B.; Álvarez, A.; Bae, S. Hydrothermal Carbonization: Modeling, Final Properties Design and Applications: A Review. *Energies* **2018**, *11*, 216, doi:10.3390/en11010216.

138. Thorsness, C.B. *Process modeling of hydrogen production from municipal solid waste*; Livermore, CA, 1995;

139. Channiwala, S.A.; Parikh, P.P. A unified correlation for estimating HHV of solid, liquid and gaseous fuels. *Fuel* **2002**, *81*, 1051–1063, doi:10.1016/S0016-2361(01)00131-4.

140. Kieseler, S.; Neubauer, Y.; Zobel, N. Ultimate and Proximate Correlations for Estimating the Higher Heating Value of Hydrothermal Solids. *Energy & Fuels* **2013**, *27*, 908–918, doi:10.1021/ef301752d.

141. Gerber, M.; Schneider, N. Density of biogas digestate depending on temperature and composition. *Bioresour. Technol.* **2015**, *192*, 172–176, doi:10.1016/j.biortech.2015.05.061.

142. Met Office UK climate Available online: http://www.metoffice.gov.uk/public/weather/climate/ (accessed on Aug 16, 2017).

143. Furukawa, H.; Kato, Y.; Inoue, Y.; Kato, T.; Tada, Y.; Hashimoto, S. Correlation of Power Consumption for Several Kinds of Mixing Impellers. *Int. J. Chem. Eng.* **2012**, *2012*, 1–6, doi:10.1155/2012/106496.

144. Tian, L.; Shen, F.; Yuan, H.; Zou, D.; Liu, Y.; Zhu, B.; Li, X. Reducing agitation energy-consumption by improving rheological properties of corn stover substrate in anaerobic digestion. *Bioresour. Technol.* **2014**, *168*, 86–91, doi:10.1016/j.biortech.2014.03.023.

145. Metzner, A.B.; Feehs, R.H.; Ramos, H.L.; Otto, R.E.; Tuthill, J.D. Agitation of viscous Newtonian and non-Newtonian fluids. *AIChE J.* **1961**, *7*, 3–9, doi:10.1002/aic.690070103.

146. Mbaye, S.; Dieudé-Fauvel, E.; Baudez, J.C. Comparative analysis of anaerobically digested wastes flow properties. *Waste Manag.* **2014**, *34*, 2057–2062, doi:10.1016/j.wasman.2014.06.021.

147. Huwaldt, J.A.; Steinhorst, S. Plot Digitizer Available online: http://plotdigitizer.sourceforge.net/ (accessed on Jun 27, 2017).

148. S, P.; Perry, R.H.; Green, D.W.; O Maloney, J. *Chemical Engineers' Handbook Seventh Edition*; 1997; Vol. 27; ISBN 0070498415.

149. Towler, G.; Sinnott, R. Chapter 15 - Design of Reactors and Mixers. In *Chemical Engineering Design*; Elsevier, 2013; pp. 631–751.

150. ASME *BPVC Section VIII-Rules for Construction of Pressure Vessels Division 1*; ASME, 2019; ISBN 9780791872871.

151. Corma, A.; Renz, M. Chapter 2: Chemistry of the HTC process and technology. In *NEWAPP - Industrial Scale Hydrothermal Carbonization: new applications for wet biomass waste*; 2016; p. 5.

152. Woods, D.R. Appendix D: Capital Cost Guidelines. *Rules Thumb Eng. Pract.* 2007.

153. Harker, J.H.; Backhurst, J.R.; Richardson, J.F. 16.5.8. Fluidised bed dryers. In *Coulson and Richardson's Chemical Engineering Volume 2 - Particle Technology and Separation Processes*; 2002 ISBN 9780750644457.

154. Seider, W.D.; Seader, J.D.; Lewin, D.R.; Widagdo, S.; Gani, R.; Ng, K.M. *Product and Process Design Principles: Synthesis, Analysis and Design*; John Wiley & Sons, 2017; ISBN 978-0470048955.

155. Towler, G.; Sinnott, R. Chapter 7 - Capital Cost Estimating. In *Chemical Engineering Design*; Towler, G., Sinnott, R.B., Eds.; Butterworth-Heinemann: Boston, 2013; pp. 307–354 ISBN 978-0-08-096659-5.

156. Chemical Engineering *Chemical Engineering Plant Cost Index: 2019 Annual Value*; 2019;

157. X-Rates British Pound per 1 US Dollar Monthly average Available online: https://www.x-rates.com/average/?from=USD&to=GBP&amount=1&year=2018 (accessed on Jun 12, 2020).

158. X-Rates British Pound per 1 Euro Monthly average Available online: https://www.x-rates.com/average/?from=EUR&to=GBP&amount=1&year=2019 (accessed on Jun 12, 2020).

159. Eurostat Harmonised index of consumer prices (HCIP) 2020.

160. Intratec Plant Construction Index for Cost Escalation Available online: https://www.intratec.us/indexes-and-pricing-data/plant-construction-index (accessed on Feb 4, 2019).

161. Ulrich, G.D.; Vasude, P.T. *Chemical Engineering Process Design and Economics: A Practical Guide*; 2nd Editio.; Process Publishing, 2004; ISBN 0970876823.

162. Towler, G.; Sinnott, R. Chapter 8 - Estimating Revenues and Production Costs. In *Chemical Engineering Design*; Towler, G., Sinnott, R.B., Eds.; Butterworth-Heinemann: Boston, 2013; pp. 355–387.

163. BEIS *Prices of fuels purchased by non-domestic consumers in the UK*; 2020;

164. BSI *PAS 110:2010 Specification for whole digestate, separated liquor and separated fibre derived from the anaerobic digestion of source-segregated biodegradable materials*; 2010;

165. Office for National Statistics *Earnings and hours worked, occupation by four-digit SOC: ASHE Table 14*; 2018;

166. European Commission *Survey on the Access to Finance of Enterprises (SAFE)*; 2018;

167. AHDB *GB Fertiliser Price Market Update*; 2020;

168. CCUS Cost Challenge Taskforce *Delivering Clean Growth: CCUS Cost Challenge Taskforce Report*; 2018;

169. NFU Cymru NFU Cymru astonished at publishing of draft NVZ regulations during Covid-19 crisis Available online: https://www.nfu-cymru.org.uk/news/latest-news/nfu-cymru-astonished-at-publishing-of-draft-nvz-regulations-during-covid-19-crisis/ (accessed on May 18, 2020).

170. UK Biochar Research Centre *Biochar Quality Mandate*; 2013;

171. ADBA AD Map Available online: http://adbioresources.org/map.

172. WRAP *Food Surplus and Waste in the UK – key facts*; 2020;

173. DEFRA *The Guide to Cross Compliance in England 2018*; 2018;

174. Cao, Z.; Jung, D.; Olszewski, M.P.; Arauzo, P.J.; Kruse, A. Hydrothermal carbonization of biogas digestate: Effect of digestate origin and process conditions. *Waste Manag.* **2019**, *100*, 138–150, doi:10.1016/j.wasman.2019.09.009.

175. Lucian, M.; Volpe, M.; Merzari, F.; Wüst, D.; Kruse, A.; Andreottola, G.; Fiori, L. Hydrothermal carbonization coupled with anaerobic digestion for the valorization of the organic fraction of municipal solid waste. *Bioresour. Technol.* **2020**, *314*, 123734, doi:10.1016/j.biortech.2020.123734.

176. Mumme, J.; Titirici, M.M.; Pfeiffer, A.; Lüder, U.; Reza, M.T.; Mašek, O. Hydrothermal Carbonization of Digestate in the Presence of Zeolite: Process Efficiency and Composite Properties. *ACS Sustain. Chem. Eng.* **2015**, *3*, 2967–2974, doi:10.1021/acssuschemeng.5b00943.

177. Rodriguez Correa, C.; Bernardo, M.; Ribeiro, R.P.P.L.; Esteves, I.A.A.C.; Kruse, A. Evaluation of hydrothermal carbonization as a preliminary step for the production of functional materials from biogas digestate. *J. Anal. Appl. Pyrolysis* **2017**, *124*, 461–474, doi:10.1016/j.jaap.2017.02.014.

178. Reza, M.T.; Mumme, J.; Ebert, A. Characterization of hydrochar obtained from hydrothermal carbonization of wheat straw digestate. *Biomass Convers. Biorefinery* **2015**, *5*, 425–435, doi:10.1007/s13399-015-0163-9.

179. Zhang, D.; Wang, F.; Shen, X.; Yi, W.; Li, Z.; Li, Y.; Tian, C. Comparison study on fuel properties of hydrochars produced from corn stalk and corn stalk digestate. *Energy* **2018**, *165*, 527–536, doi:10.1016/j.energy.2018.09.174.

180. Funke, A. Fate of plant available nutrients during hydrothermal carbonization of digestate. *Chemie-Ingenieur-Technik* **2015**, *87*, 1713–1719, doi:10.1002/cite.201400182.

181. Tillman, D.A.; Duong, D.N.B.; Harding, N.S. *Solid Fuel Blending - Principles, Practices, and Problems*; Elsevier, 2012; ISBN 9780123809322.

182. Ingelia Ingelia Model Available online: https://ingelia.com/index.php/negocio-sostenible/modelo-ingelia/?lang=en (accessed on Sep 24, 2020).

183. Oliver-Tomas, B.; Hitzl, M.; Owsianiak, M.; Renz, M. Evaluation of hydrothermal carbonization in urban mining for the recovery of phosphorus from the organic fraction of municipal solid waste. *Resour. Conserv. Recycl.* **2019**, *147*, 111–118, doi:10.1016/j.resconrec.2019.04.023.

184. BEIS *Greenhouse gas reporting: conversion factors 2019*; 2019;

185. Eurostat Labour cost levels by NACE Rev. 2 activity Available online:

https://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=lc_lci_lev&lang=en.

186. Stirling, R.J. An investigation into the impact of hydrothermal carbonisation on the suitability of biomass for fuel switching, The University of Nottingham, 2019.

187. Pawlak-Kruczek, H.; Niedzwiecki, L.; Sieradzka, M.; Mlonka-Mędrala, A.; Baranowski, M.; Serafin-Tkaczuk, M.; Magdziarz, A. Hydrothermal carbonization of agricultural and municipal solid waste digestates – Structure and energetic properties of the solid products. *Fuel* **2020**, *275*, doi:10.1016/j.fuel.2020.117837.

188. Which? Three food waste facts everyone needs to know Available online: https://www.which.co.uk/news/2019/06/three-food-waste-facts-everyone-needs-to-know/.

189. HM Revenue & Customs Landfill Tax rates Available online: https://www.gov.uk/government/publications/rates-and-allowances-landfill-tax/landfill-tax-rates-from-1-april-2013 (accessed on Sep 16, 2020).

190. WRAP *Gate Fees 2018/19 Report: Comparing the cost of alternative waste treatment options*; 2019;

191. Google Google Maps Available online: https://www.google.com/maps.

192. Aqua Enviro *Digestate Dewatering: Commercial State of the Art*; 2016;

193. Environment Agency *Nitrate Vulnerable Zones Map*; 2019;

194. NERA Economic Consulting *Electricity Generation Costs and Hurdle Rates, Lot 1: Hurdle Rates update for Generation Technologies*; 2015;

195. Forest Research Woodfuel economics Available online: https://www.forestresearch.gov.uk/tools-and-resources/urban-regeneration-and-greenspace-partnership/greenspace-in-practice/benefits-of-greenspace/woodfuel-economics/.

196. CALU *Economics of Miscanthus and SRC production*; 2006;

197. Von Massow, H.-J.; Wittmann, T. Method and device for hydrothermal carbonization (HTC) of biomass using an htc system 2008.

Appendix

Table A.1: Mass yield and proximate analysis results. daf represents a dry, ash-free basis. VM is the volatile matter and FC is the fixed carbon.

	Run order	Experimental conditions			Mass Yield (wt. % raw)			Mass Yield (wt. % daf)			Proximate (wt. % dry)		
		Temp (°C)	Time (min)	Mois. (%)	Solid	Liquid	Gas	Solid	Liquid	Gas	VM	FC	Ash
Waste-based	1	215	120	87.5%	52.7%	43.9%	3.4%	41.9%	52.6%	5.4%	49.1%	1.1%	49.8%
	2	180	120	80.0%	55.3%	43.8%	0.9%	46.8%	51.8%	1.4%	52.7%	1.0%	46.3%
	3	215	120	87.5%	51.7%	46.1%	2.2%	40.7%	55.8%	3.5%	50.2%	-0.4%	50.3%
	4	215	60	95.0%	50.0%	49.5%	0.6%	39.7%	59.4%	0.9%	49.7%	0.5%	49.8%
	5	215	180	95.0%	50.0%	49.3%	0.7%	39.3%	59.5%	1.2%	49.3%	0.2%	50.5%
	6	250	180	87.5%	49.7%	46.6%	3.8%	38.8%	55.2%	6.0%	49.9%	-0.9%	51.0%
	7	250	120	95.0%	48.7%	49.6%	1.7%	35.1%	62.2%	2.7%	45.6%	-0.4%	54.8%
	8	215	120	87.5%	51.7%	46.8%	1.6%	42.8%	54.7%	2.5%	50.5%	1.6%	47.9%
	9	215	180	80.0%	51.7%	43.6%	4.8%	40.7%	51.7%	7.7%	49.3%	0.3%	50.4%
	10	215	60	80.0%	51.3%	45.9%	2.8%	42.0%	53.5%	4.5%	51.3%	0.5%	48.2%
	11	250	60	87.5%	51.0%	44.6%	4.4%	39.9%	53.1%	7.1%	48.3%	0.9%	50.8%
	12	250	120	80.0%	52.8%	40.9%	6.3%	40.8%	49.1%	10.1%	47.5%	1.1%	51.4%
	13	180	180	87.5%	57.0%	41.2%	1.8%	47.6%	49.6%	2.8%	51.7%	1.3%	47.0%
	14	180	120	95.0%	55.7%	43.8%	0.5%	45.2%	54.0%	0.8%	50.8%	0.8%	48.4%
	15	180	60	87.5%	57.7%	41.5%	0.8%	48.6%	50.1%	1.3%	52.0%	1.6%	46.4%
	Feedstock				-	-	-	-	-	-	59.6%	3.9%	36.5%
Farm-based	1	190	60	85	60.8%	37.7%	1.5%	59.2%	38.8%	2.0%	58.3%	14.8%	26.9%
	2	190	180	85	58.9%	38.3%	2.8%	56.4%	39.9%	3.8%	54.0%	17.7%	28.3%
	3	220	60	85	52.7%	42.7%	4.6%	50.5%	43.2%	6.3%	55.0%	16.3%	28.7%
	4	220	180	85	49.8%	44.3%	5.9%	47.8%	44.1%	8.0%	52.3%	19.1%	28.7%
	5	250	60	85	53.5%	39.3%	7.2%	49.6%	40.5%	9.8%	50.2%	18.8%	31.0%
	6	250	180	85	45.6%	46.3%	8.2%	41.4%	47.5%	11.1%	46.9%	20.1%	32.9%
	Feedstock				-	-	-	-	-	-	59.6%	17.4%	22.9%
Crop-based	1	180	60	88.9%	68.3%	-	-	72.1%	-	-	80.9%	13.3%	5.8%
	2	200	60	88.9%	68.3%	-	-	70.6%	-	-	71.1%	19.6%	9.4%
	3	215	60	93.8%	60.0%	-	-	62.7%	-	-	74.9%	17.7%	7.4%
	4	215	60	75.0%	64.2%	-	-	67.5%	-	-	72.1%	20.5%	7.5%
	5	215	90	88.9%	58.3%	-	-	62.0%	-	-	73.4%	20.1%	6.7%
	6	250	60	88.9%	42.7%	-	-	43.0%	-	-	56.5%	31.4%	12.1%
	Feedstock				-	-	-	-	-	-	70.4%	20.2%	9.3%

Table A.2: Elemental composition of the hydrochars and feedstocks on a dry and dry, ash-free (daf) basis. The oxygen content was calculated by difference.

	Run order	Experimental conditions			Elemental composition (wt. % dry)				Elemental composition (wt.% daf)			
		Temp (°C)	Time (min)	Mois. (%)	C	H	N	O	C	H	N	O
Waste-based	1	215	120	87.5%	27.3%	3.5%	1.5%	17.3%	54.4%	7.0%	3.0%	34.5%
	2	180	120	80.0%	28.5%	3.2%	2.0%	19.0%	53.2%	6.0%	3.6%	35.5%
	3	215	120	87.5%	28.8%	3.2%	1.7%	15.4%	57.9%	6.4%	3.3%	31.0%
	4	215	60	95.0%	27.2%	3.0%	1.4%	17.9%	54.2%	6.0%	2.8%	35.7%
	5	215	180	95.0%	26.8%	2.9%	1.2%	18.1%	54.0%	5.9%	2.5%	36.6%
	6	250	180	87.5%	28.7%	3.0%	1.5%	15.5%	58.6%	6.2%	3.0%	31.5%
	7	250	120	95.0%	24.9%	2.6%	0.9%	16.5%	55.0%	5.7%	2.1%	36.5%
	8	215	120	87.5%	29.5%	3.2%	1.8%	17.2%	56.6%	6.1%	3.4%	33.0%
	9	215	180	80.0%	27.0%	2.7%	1.5%	17.7%	54.5%	5.5%	3.1%	35.8%
	10	215	60	80.0%	28.4%	3.0%	1.7%	17.9%	54.8%	5.7%	3.3%	34.6%
	11	250	60	87.5%	27.1%	2.8%	1.4%	17.4%	55.0%	5.7%	2.8%	35.3%
	12	250	120	80.0%	26.4%	2.6%	1.4%	17.8%	54.2%	5.4%	2.9%	36.6%
	13	180	180	87.5%	26.5%	2.8%	1.7%	21.1%	49.9%	5.3%	3.2%	39.8%
	14	180	120	95.0%	27.2%	2.9%	1.8%	18.7%	52.8%	5.5%	3.5%	36.2%
	15	180	60	87.5%	27.8%	3.0%	2.0%	19.8%	51.9%	5.5%	3.6%	36.9%
	Feedstock				27.6%	3.8%	3.5%	29.4%	43.5%	6.0%	5.5%	46.4%
Farm-based	1	190	60	85	42.8%	4.9%	3.0%	21.4%	58.5%	6.7%	4.1%	29.3%
	2	190	180	85	44.2%	4.7%	2.9%	18.9%	61.7%	6.6%	4.0%	26.4%
	3	220	60	85	44.8%	5.1%	2.6%	18.1%	62.8%	7.2%	3.7%	25.4%
	4	220	180	85	46.4%	4.9%	2.7%	16.7%	65.1%	6.8%	3.8%	23.4%
	5	250	60	85	45.6%	4.8%	3.0%	14.9%	66.1%	7.0%	4.4%	21.6%
	6	250	180	85	49.5%	5.1%	3.0%	9.1%	73.8%	7.6%	4.5%	13.6%
	Feedstock				38.6%	4.6%	3.7%	33.7%	50.1%	5.9%	4.8%	43.7%
Crop-based	1	180	60	88.9%	49.2%	6.0%	1.2%	37.4%	52.2%	6.3%	1.3%	39.7%
	2	200	60	88.9%	51.5%	6.1%	1.6%	31.3%	56.8%	6.7%	1.8%	34.5%
	3	215	60	93.8%	52.2%	5.8%	1.4%	32.8%	56.4%	6.3%	1.5%	35.5%
	4	215	60	75.0%	53.2%	5.7%	1.7%	31.7%	57.5%	6.2%	1.9%	34.2%
	5	215	90	88.9%	52.9%	5.8%	1.6%	32.9%	56.7%	6.2%	1.7%	35.3%
	6	250	60	88.9%	59.6%	5.4%	2.0%	20.8%	67.8%	6.1%	2.3%	23.7%
	Feedstock				39.9%	5.0%	2.3%	46.6%	44.0%	5.5%	2.5%	51.4%

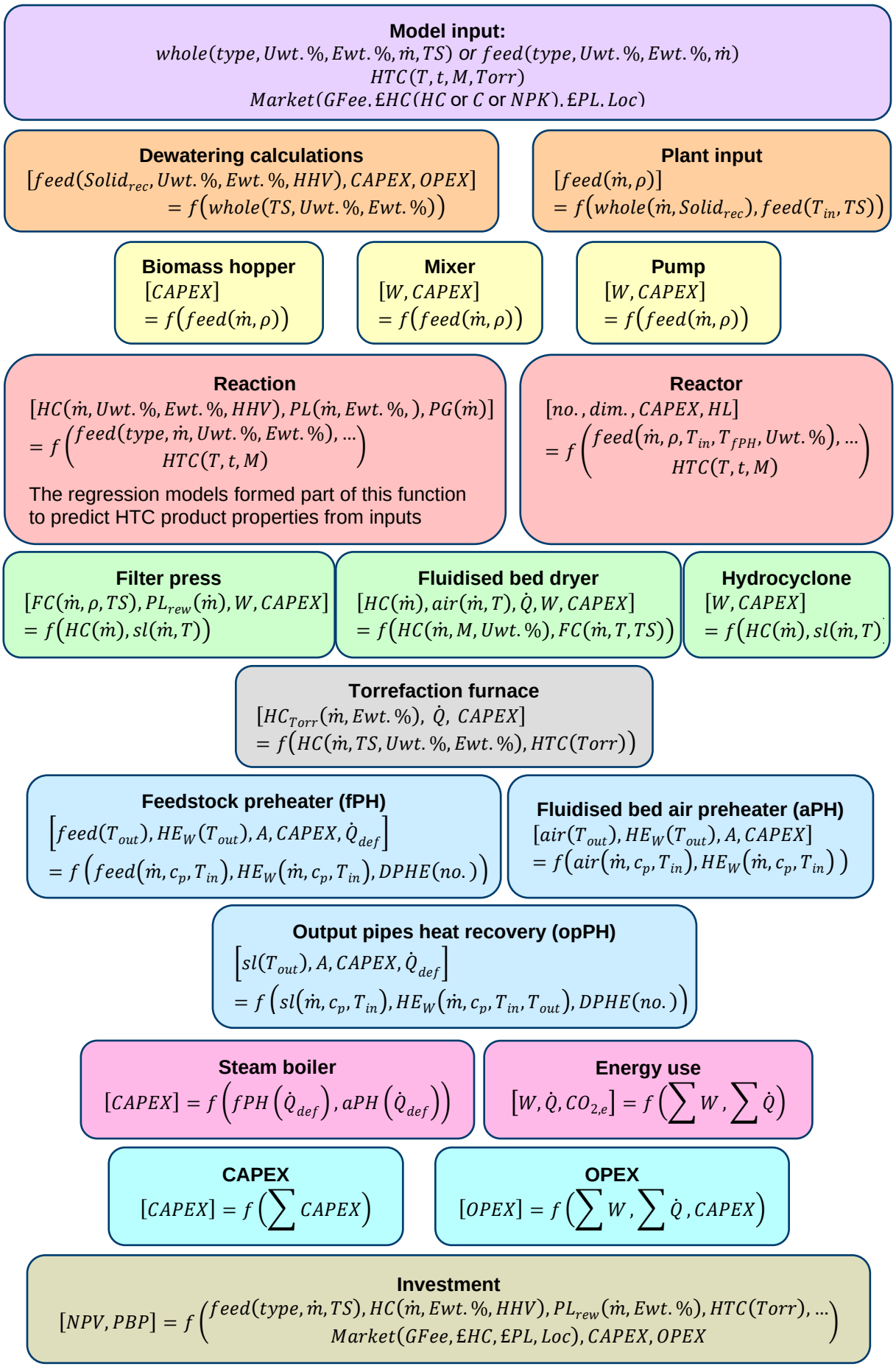


Figure A.1: The functions involved in the process model whereby $[output(s)] = f(input(s))$ and notation is used for the subject and properties e.g., $feed(Solid_{rec}, Uwt.\%, Ewt.\%, HHV)$ represents the solid recovery, ultimate analysis, elemental analysis and high heating value of the feedstock. Definitions for these notations are shown in Table A.3 and Table A.4.

Table A.3: Definitions of the subjects used in Figure A.1.

Notation	Definition	Notation	Definition
$feed$	Feedstock	$Market$	Market conditions
$whole$	Whole digestate	air	Air in fluidised bed dryer
HC	Hydrochar	HE_W	Heat exchange network water
PL	Process liquid	fPH	Feedstock preheater
PG	Process gas	$opPH$	Output pipes heat recovery
sl	Slurry (products post HTC reactor)	aPH	Fluidised bed air preheater
PL_{rew}	Recovered process liquid	$DPHE$	Double-pipe heat exchanger (fPH & opPH)
FC	Feedcake (products post-filter press)		
HC_{Torr}	Torrefied hydrochar		

Table A.4: Definitions of the properties used in Figure A.1.

Notation	Definition	Notation	Definition
$type$	Type of feedstock (waste-based, farm-based etc.)	$CAPEX$	CAPEX
$\dot{m}$	Mass flowrate	$OPEX$	OPEX
TS	Total solids	W	Electrical power
ρ	Density	$\dot{Q}$	Heat
$Uwt.\%,$	Ultimate analysis (VM, FC, Ash)	$\dot{Q}_{def}$	Heat deficit
$Ewt.\%,$	Elemental analysis (C, H, N, O, P, K)	$no.$	Number of units
HHV	High heating value	$dim.$	Dimensions
T	Temperature	HL	Heat loss
t	Residence time	A	Heat exchanger area
M	Moisture content	c_p	Specific heat capacity
$Torr$	Torrefaction post-treatment (on/off)	ΔT	Temperature difference
$GFee$	Gate fee	U	Overall heat transfer coefficient
$£HC$	Hydrochar sale price	$CO_{2,e}$	CO_2 equivalent
$£PL$	Process liquid sale price	NPV	Net present value
Loc	Location (Notts/Lincs/neither)	PBP	Payback period
$Solid_{rec}$	Solid recovery		